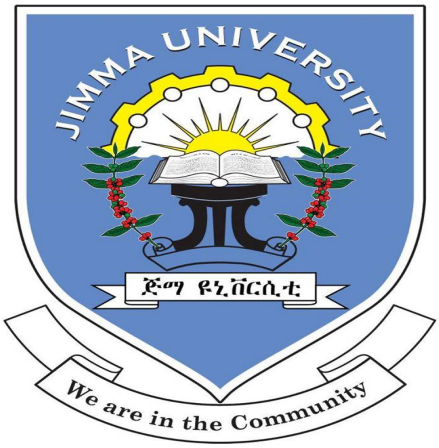

THIRD ORDER NONLINEAR INDEX OF REFRACTION AND ABSORPTION COEFFICIENT OF ZINC OXIDE QUANTUM DOT

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A Thesis Submitted to

The Department of Physics

PRESENTED IN FULFILMENT OF THE
REQUIREMENTS FOR THE DEGREE OF MASTER OF SCIENCE
JIMMA UNIVERSITY
JIMMA, ETHIOPIA
OCTOBER 2017

JIMMA UNIVERSITY
DEPARTMENT OF
PHYSICS

This is to certify that the thesis prepared by **Alemayehu Fulas** Graduate Studies entitled "**third order nonlinear index of refraction and absorption coefficient of Zinc Oxide quantum dot**" in fulfillment of the requirements for the degree of **Master of Science** complies with the regulations of the University and meets the accepted standards with respect to originality and quality. .

Dated: October 2017

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JIMMA UNIVERSITY

Date: **October 2017**

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Title: **third order nonlinear index of refraction and absorption coefficient of Zinc Oxide quantum dot**

Department: **Physics**

Degree: **M.Sc.** Convocation: **October** Year: **2017**

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To my son

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Abstract

In this research analytical and numerical calculations have been made to identify the dependence of third order nonlinear refractive index and absorption coefficient on optical laser intensity and transition energy. Analytical expressions for the third-order nonlinear absorption coefficients and refractive index changes are obtained by using compact density matrix approach. The obtained result indicates that both third order NL absorption coefficient and refractive index changes experience a blue shift by increasing the transition energy. Moreover high laser intensity induces a pronounced magnitude of third order NL absorption coefficient and refractive index changes.

Acknowledgements

First, I would like to thank the Almighty GOD. It is only due to His blessings that I have been able to accomplish this work. I express my deep sense of gratitude to my advisor Dr. Menberu Mengesha for his excellent guidance, competent advice. I am deeply thankful to him for his kindness, constant encouragement and support, without which the successful completion of this work would not have been possible. I would like to thank my co-advisor, Mr Hiwot Digafe, for his support while carrying out this research work. I extend my sincere thanks to instructors; Mr Chali Yadeta ,Mr Solomon H/Mariam and all staff members in the physics department. It is with a particular pleasure that I acknowledge my friend Endal Meseret for his valuable suggestions and discussions throughout this work, he make me strong. I would like to express my heart full thanks and love to my wife and my son Fitsum, my siters and without their help this work for their support through out this work.would not have been possible.Finally I would like to thank the Department of Physics for facilitating all the necessary materials while I am conducting my research and I am thankful to graduate school of Jimma University for they gave me this chance and their economical support.

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Chapter 1

Introduction

1.1 Background of the Study

What is **NONLINEAR OPTICS** ?

The ordinary experiences of everyday life *reflection, refraction, diffraction, absorption*, e.t.c. explain a wide variety of common visual experiences, which are independent of the intensity of light. Such optical phenomena is the realm of the so called **linear optics**[11].

Nonlinear optics is *a field of study concerned with the interaction of intense electromagnetic radiation and matter in which the matter responds in a nonlinear manner to the incident optical fields.* In other words, nonlinear optics (NLO) is the branch of optics that describes the behavior of light in nonlinear media, in which the polarization P responds nonlinearly to the electric field E of the light[1,11]. This nonlinearity is typically observed at very high light intensities, such as those provided by lasers. Such high powers of laser beams made it possible, for the first time, to observe that the effect of light on a medium can indeed change its optical properties, such as refractive index and absorption coefficient.

With the advent of the laser in 1960, nonlinear optics went from the realm of

theoretical prediction to experimental reality, and it becomes a rapidly growing field of research in recent decades. In fact, the beginning of the field of nonlinear optics is taken to be the discovery of second-harmonic generation by Fraken et al. (1961), shortly after the demonstration of the first working laser by Maiman in 1960 [1,4] and first observation of third harmonic generation (THG) Terhuneetal.(1962) [3].

Nonlinear optical phenomena are "nonlinear" in a 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 [1,11].

In linear optics, light traveling through media induces an oscillating polarization that is linear with its electromagnetic field. In nonlinear optics, coherent light can distort the oscillating polarization; these distortions can create or enhance new fields that then radiate from the oscillating nonlinear polarization [2].

This nonlinearity is highly pronounced in nanoparticles, 500 times their bulk counterpart [17]. Nanoscale materials are under active research over the past few decades, owing to their interesting versatile properties, quite different from those of the bulk form [6,8].Thus the nonlinear optical properties of nanostructured semiconductors are the topics of current theoretical and experimental interest,[5,9,10]. Amongst the various NLO materials investigated, direct band-gap semiconductors, such as zinc oxide have attractive nonlinear properties that make them ideal candidates for NLO based devices.

Some of the potential applications of oxide semiconductor nanocrystals are in *field emission displays, solar cells, gas sensors, photovoltaic, display devices and laser fabrication* [7-9, 11]. The refractive index of a material is the key parameter that affects all optical properties. Any modification of the refractive index, leads to new optical properties of absorption and dispersion of the medium [14].

1.2 Statement of the problem

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Recently the nonlinear optical response of nanoparticles (ZnO) is greatly investigated theoretically and experimentally in relation with confining energy, impurity,e.t.c. But, up to our knowledge there is no studies concerning ZnO spherical quantum dot in density matrix formalism. What are the effects of transition energy and optical laser intensity on NL-RIC and AC of spherical QD ?

1.3 Objectives

1.3.1 General objective

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Mainly our research work focused on the third-order nonlinear optical phenomena and nonlinear optical properties of ZnO quantum dot. We determined the effect of transition energy and optical intensity on third order nonlinear refractive index changes and absorption coefficient of ZnO quantum dot.

1.3.2 Specific objective

In this theoretical research the following specific works are done:

- We calculated the transition energy of ZnO quantum dot.
- Analytically we developed equation of third order NL susceptibility.
- Numerical calculations of third order NL absorption coefficient and refractive index changes of ZnO quantum dot was performed.

1.4 Significance of the Study

ZnO has emerged as one of the promising materials due to its unique properties such as high mechanical and chemical stability, excellent electrical and optical properties together with its natural abundance and non-toxicity [13]. ZnO thin films (QD) have potential technological application. Most of its potentials arises from it's nonlinear optical property, which is basically described by the nonlinear refractive index and absorption coefficients. We believe that our results can be useful and helpful in providing some additional knowledge to the scientific society and researchers in relation to third order nonlinear ; refractive index and absorption coefficient of ZnO quantum dot.

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Chapter 2

Literature Review

2.1 Nonlinear Optics and Nanoparticles

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Nanomaterial,that exhibit peculiar properties which are not shown by their bulk counterparts, have attracted much interest from both fundamental and technological researchers. Among the various nonlinear optical (NLO) materials investigated, wide band gap semiconductors, especially zinc oxide (ZnO), have attractive nonlinear properties that make them ideal candidates for NLO-based devices [9]. Nano sized ZnO in the form of quantum dots, nanowires, Nano-belts, etc. are referred to as the material of the 21st century [23].

2.1.1 Nonlinear Polarization and Susceptibility

. The most usual procedure for describing nonlinear optical phenomena is based on expressing the polarization P(t) in terms of the applied electric field strength E(t). The nonlinear susceptibility is a quantity that is used to determine the nonlinear polarization of a material medium in terms of the strength of an applied optical - frequency electric field. It thus provides a framework for describing nonlinear optical

phenomena: such as, nonlinear refraction and absorption coefficient [1]. In case of linear optics, the induced polarization depends linearly on the electric field strength in a manner that can often be described by the relationship;

$$P(t) = \epsilon_0 \chi^{(1)} E(t) \tag{2.1.1}$$

where the constant of proportionality $\chi^{(1)}$is known as the linear susceptibility and ϵ_0 is the permittivity of free space. In nonlinear optics, the optical response can often be described by generalizing Eq.(2.1.1) and expressing the polarization P(t) as a power series in the field strength E(t) [1,11]

$$\begin{aligned} P(t) &= \epsilon_0[\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + ...] \\ &= P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + ... \end{aligned} \tag{2.1.2}$$

Where, $\chi^{(2)}$ *and* $\chi^{(3)}$ are the second- and third-order nonlinear optical susceptibilities, respectively. The nonlinear polarization given by Eq.(2.1.2) is only for a material system that is lossless and dispersionless. In general, the nonlinear susceptibilities depend on the frequencies of the applied fields. Third-order optical nonlinearities cover a vast and diverse area in nonlinear optics[12]. The contribution of third order nonlinear polarization is

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t) \tag{2.1.3}$$

Which is induced by an applied field $\vec{E}(t)$ that consists of three frequency component

$$\vec{E}(t) = E_1 e^{(-i\omega_1 t)} + E_2 e^{(-i\omega_2 t)} + E_3 e^{(-i\omega_3 t)} + c.c. \tag{2.1.4}$$

Where, c.c. is the complex conjugate of the electric field vector. For the simple case in which the applied field is monochromatic, E(t)is given by

$$\vec{E}(t) = E \cos \omega t \tag{2.1.5}$$

Then, through use of trigonometry identity,

$$\cos^3 \omega t = \frac{1}{4}\cos 3\omega t + \frac{3}{4}\cos \omega t$$

we can express the third order nonlinear polarization as

$$P^{(3)}(t) = \epsilon_0 E^3 \frac{1}{4}\cos 3\omega t + \epsilon_0 E^3 \frac{3}{4}\cos \omega t \tag{2.1.6}$$

The two terms in the above equation(eq.2.1.6) has their own descriptions. The first term, a response at frequency 3ω that is created by an applied field of frequency ω is the nonlinear contribution to the polarization at the frequency of the incident field. Hence it is the nonlinear contribution of the polarization to the nonlinear index of refraction [1]. The index of refraction in the presence of this type of nonlinearity can be represented as

$$n = n_0 + n_2 I \tag{2.1.7}$$

Where n_0 is the linear index of refraction and n_2 is an optical constant that characterizes the strength of the optical nonlinearity, and where, $I = \frac{2n_r}{\mu c}|E(\omega)|^2$, is the intensity of the incident wave.

2.1.2 Nonlinear Index of Refraction and Absorption Coefficient

Nonlinear Index of Refraction

The refractive index of a material is the key parameter that affects many optical properties. Many of the interesting phenomena of nonlinear optics derive their behavior from the nonlinear index of refraction. which led to a variety of fascinating applications. It is also central to many fundamental scientific investigations.Even though several diverse physical effects contribute to the nonlinear index of refraction,

generally, it can be defined a[11]s a change in the refractive index or the spatial distribution of the refractive index of a medium due to the presence of optical waves. The nonlinear index of refraction has generated significant scientific and technological interest and it has been utilized in or considered for a variety of applications, such as: *Nonlinear spectroscopy, Correcting optical distortions, Opticalswitching, Optical logic gates, Optical data processing, Optical communications, Optical limiting, Passive laser mode-locking, Wave guide switches and modulators*. These applications derive from several phenomena that have been discovered and are related to the nonlinear refractive index [11,12].

Nonlinear Absorption

Refers to the change in transmittance of a material as a function of intensity. At sufficiently high intensities, the probability of a material absorbing more than one photon before relaxing to the ground state can be greatly enhanced. The many different effects produced by nonlinear absorption in the frequency dependent transmittance of a material have led to several applications in science and technology, such as *nonlinear spectroscopy and optical limiting* [11,12]. The nonlinear refractive index n_2 is related to the real part of nonlinear susceptibility$[Re\chi^{(3)}]$ and the imaginary part of third order susceptibility $[I_m\chi^{(3)}]$ determines the strength of the nonlinear absorption[9,16,21,27].

$$\frac{\Delta n(\omega)}{n_r} = \frac{1}{2n_r^2} R_e\chi(\omega) \tag{2.1.8}$$

The nonlinear absorption coefficient,β is related to $I_m\chi^{(3)}$ by the relation,

$$\beta^{(3)}(\omega) = \frac{\omega}{n_r}\sqrt{\frac{\mu}{\epsilon_0}} I_m\chi^{(3)}(\omega) \tag{2.1.9}$$

Where n_r is the refractive index of the medium, ϵ_0 is the permittivity of free space and c is the velocity of light in vacuum. From the real and imaginary parts of $\chi^{(3)}$,

the modulus of third order nonlinear susceptibility can be found out using the above two equations.

$$| \chi^{(3)} | = [| R_e\chi^{(3)} | + | I_m\chi^{(3)} |]^{\frac{1}{2}} \tag{2.1.10}$$

The magnitude of $(\chi^{(3)})$ is significantly affected by the crystallite size and it determines the strength of nonlinearity of the material [11].

2.2 Maxwell's Equation of Electromagnetic Wave

Light is an electromagnetic wave. It consists of electric and magnetic fields, E and H. For most of optics, the optical wave may be characterized by defining its electric field. The magnetic field is related to the electric field through Maxwell's equations. The nonlinearity in the response of a material system to an intense laser field can cause the polarization of the medium to develop new frequency components not present in the incident radiation field. These new frequency components of the polarization act as sources of new frequency components of the electromagnetic field. Thus, Maxwell's electromagnetic wave equations describe these phenomena. The general forms of Maxwell's equations of electromagnetic waves are described by the following four equations [1,18];

$$\vec{\nabla}.\vec{D} = \vec{\rho} \tag{2.2.1}$$

$$\vec{\nabla}.\vec{B} = 0 \tag{2.2.2}$$

$$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \tag{2.2.3}$$

$$\vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t} + \vec{J} \tag{2.2.4}$$

Where E is electric field, D is electric displacement, H is magnetic field intensity, B is magnetic induction, $\vec{\rho}$ is the electric charge density and J is the electric current density. In regions of space where there are no free charges and no free current, we have

$$\vec{\rho} = 0$$

and

$$\vec{J} = 0$$

Taking the assumptions that the material is nonmagnetic and nonlinear, we can use the relations

$$\vec{B} = \mu_0 \vec{H} \; and \; \vec{D} = \mu_0 \vec{E} + \vec{P} \tag{2.2.5}$$

$\vec{P}$ is the total polarization. So that

$$\vec{P} = \vec{P}^{L} + \vec{P}^{NL}$$

2.2.1 Electromagnetic Wave Equation of NLO

After some substitutions and operations of equations (2.2.1) to (2.2.4), we obtain Maxwell's optical wave equation for nonlinear medium, which can be written as [1,11]:

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} + \mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2} = 0 \tag{2.2.6}$$

By substituting $\mu_0 \epsilon_0 = \frac{1}{c^2}$, we obtain

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} + \frac{\partial^2 \vec{E}}{c^2 \partial t^2} = -\frac{\partial^2 \vec{P}}{\epsilon_0 c^2 \partial t^2} = 0 \tag{2.2.7}$$

From vetor identity we know that

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} = \vec{\nabla}(\vec{\nabla}.\vec{E}) - \vec{\nabla}^2\vec{E}$$

But the first term of the right hand side of the identity equation is either vanish or its contribution is negligible, thus the wave equation will reduced to

$$\vec{\nabla}^2\vec{E} - \frac{\partial^2\vec{E}}{c^2\partial t^2} = \frac{\partial^2\vec{P}}{\epsilon_0 c^2\partial t^2} \tag{2.2.8}$$

For simple case of an *isotropic, dispersionless* material, the wave equation becomes

$$-\vec{\nabla}^2\vec{E} + \frac{\epsilon^{(1)}\partial^2\vec{E}}{c^2\partial t^2} = -\frac{\partial^2\vec{P}^{(NL)}}{\epsilon_0 c^2\partial t^2} \tag{2.2.9}$$

Where $\epsilon^{(1)}$ is relative permittivity, a dimensionless scalar quantity, which is different for each material [1]. The nonlinear response of the medium acts as a source term which appears on the right-hand side of the equation. Equation (2.2.9) can developed to an equation which is valid for each frequency component of the field.

$$-\vec{\nabla}^2\vec{E}_n + \frac{\epsilon^{(1)}(\omega_n)}{c^2}\frac{\partial^2\vec{E}}{\partial t^2} = -\frac{1}{\epsilon_0 c^2}\frac{\partial^2\vec{P}_n^{(NL)}}{\partial t^2} \tag{2.2.10}$$

2.3 Nonlinear Susceptibility of a Classical Anharmonic Oscillator

The Lorentz model of the atom, which treats the atom as a harmonic oscillator, is known to provide a very good description of the linear optical properties. By allowing the possibility of a nonlinearity in the restoring force exerted on the electron, the Lorentz model can be extend to provide a description for NLO. Though the classical model of optical nonlinearities presented here has its own shortcoming, however, it provides a good description for those cases in which all of the optical frequencies are considerably smaller than the lowest electronic resonance frequency of the material

system [1]. For the case of noncentrosymmetric media, we take the equation of motion of the electron position $\mathbf{x}$ to be of the form

$$m(\ddot{\mathbf{x}}) + 2\gamma\dot{\mathbf{x}} + \omega_0^2\mathbf{x} + ax^2) = -e\vec{E}(t) \tag{2.3.1}$$

The amplitude of the polarization component oscillating at frequency ω_q is given in terms of this amplitude by

$$\mathbf{P}^3(\omega_q) = -Ne\mathbf{r}^{(3)}(\omega_q) \tag{2.3.2}$$

Where,

$$\mathbf{r}^{(3)}(t) = \sum_q \mathbf{r}^{(3)}\omega_q e^{-i\omega_q t} \tag{2.3.3}$$

Recalling the definition of third-order nonlinear susceptibility, the polarization will be

$$P_i^{(3)}\omega_q = \sum_{ijk}\sum_{mnp}\chi_{(ijkl)}^{(3)}(\omega_q, \omega_m, \omega_n, \omega_p)E_j(\omega_m)E_k(\omega_n)E_l(\omega_p) \tag{2.3.4}$$

Thus the third-order nonlinear susceptibility in terms of first order susceptibility will becomes

$$\chi_{(ijkl)}^{(3)}(\omega_q, \omega_m, \omega_n, \omega_p) = \frac{bm\omega_0^3}{3N^3e^4}[\chi^{(1)}(\omega_q)\chi^{(1)}(\omega_m)\chi^{(1)}(\omega_n)\chi^{(1)}(\omega_p)]\times[\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}] \tag{2.3.5}$$

Where b is a parameter that characterizes the strength of the nonlinearity.

2.4 Quantum Mechanical Description of NLO

The quantum-mechanical theory of nonlinear optical susceptibility allows each atom to possess many energy eigenvalues. Quantum-mechanical (perturbation theory) of the atomic wave function enables as to derive an expressions and make more accurate

predictions of the nonlinear optical susceptibility. One of the fundamental assumption of quantum mechanics is that all of the properties of the atomic system can be described in terms of the atomic wave-function ψ(r,t), which is the solution to the time-dependent Schrodinger equation [1]

$$\hbar\frac{\partial\Psi}{\partial t} = \hat{H}\psi \tag{2.4.1}$$

Here $\hat{H}$ is the Hamiltonian operator

$$\hat{H} = \hat{H}_0 + \hat{V}(t) \tag{2.4.2}$$

Where $\hat{H}_0$ is Hamiltonian for a free atom and $\hat{V}(t)$ is the interaction Hamiltonian, which describes the interaction of the atom with the electromagnetic field. The interaction Hamiltonian is given as

$$\hat{V}(t) = -\hat{\mu}\cdot\vec{E}(t) \tag{2.4.3}$$

Here $\hat{\mu} = -er$ is the electric dipole moment operator. So that

$$\hbar\frac{\partial\Psi}{\partial t} = [\hat{H}_0 + \hat{V}(t)]\psi \tag{2.4.4}$$

2.4.1 Density Matrix Formulation of QM and NL Susceptibility.

Since we have done our research using density matrix formalism, in this section we develop the nonlinear optical susceptibility through use of the basic principles of quantum mechanics.

Why we use Density Matrix Formalism?

We use this formalism because it is capable of treating effects, such as collisional broadening of the atomic resonances that cannot be treated by Anharmonic Oscillatory Model or by simple theoretical formalism based on the atomic wave function.

We know that nonlinear effects become particularly large when one of the frequencies of the incident laser field, or when sums or differences of these frequencies, becomes equal to a transition frequency of the atomic system. But the above models or formalism does not allow us to describe the width of these resonances, and thus it cannot tell us how accurately we need to set the laser frequency to that of the atomic resonance. They also do not tell us how strongly the response is modified when the laser frequency lies within the width of the resonance [1]. That is why we choose Density Matrix Formalism for our research work.

Now it is time to begin by reviewing how the density matrix formalism derived from the basic laws of quantum mechanics. If a quantum-mechanical system (such as an atom) is known to be in a particular quantum-mechanical state s, we can describe all of the physical properties of the system in terms of the wave function $\psi_s(r,t)$ appropriate to this state. This wave function obeys the Schrodinger equation;

$$i\hbar\frac{\partial\Psi_s(r,t)}{\partial t} = \hat{H}\psi_s(r,t) \tag{2.4.5}$$

To determine how the wave evolves in time,we can represent the wave function of state s as;

$$\Psi_s(r,t) = \sum_n C_n^s(t)u_n(r) \tag{2.4.6}$$

The function $u_n(r)$ is energy eigen-solution of time- independent Schrodinger equation

$$\hat{H_0}u_n(r) = E_n u_n(r) \tag{2.4.7}$$

and $C_n^s(t)$ is the expansion coefficient which gives the probability amplitude that the atom, which is known to be in state s, is in energy eigenstate n at time t.

The time evolution of $\Psi_s(r,t)$ can be specified in terms of the time evolution of each of the expansion coefficient $C_n^s(t)$. To determine how these coefficients evolve in

time, let us introduce equation (2.4.6) into Schrodingers equation (2.4.5) to obtain

$$i\hbar \sum_n \frac{dC_n^s(t)}{dt} u_n(r) = \sum_n C_n^s(t) \hat{H} u_n(r) \tag{2.4.8}$$

In order to simplify this equation, we multiply each side from the left by u_m^* and integrate over all space. Using orthogonality condition the right-hand side is simplified by introducing the matrix elements of the Hamiltonian operator $\hat{H}$, defined through;

$$H_{mn} = \int u_m^*(r) \hat{H} u_n(r) d^3r \tag{2.4.9}$$

We thereby obtain the result

$$i\hbar \frac{d}{dt} C_m^s(t) = \sum_n \hat{H}_{mn} C_n^s(t) \tag{2.4.10}$$

Equation (2.4.10) is entirely equivalent to the Schrodinger equation (2.4.5), but it is written in terms of the probability amplitudes $C_n^s(t)$. According to QM postulate the expectation value of any observable quantity A, that corresponds to a Hermitian operator $\hat{A}$ is given by ;

$$\langle A \rangle = \int \psi_s^* \hat{A} \psi_s d^3r = \sum_{mn} C_m^{s*} C_n^s A_{mn} \tag{2.4.11}$$

Where A_{mn}, are the matrix elements.

As long as the initial state and the Hamiltonian operator $\hat{H}$ for the system are known, the formalism described by Eqs.(2.4.5) through (2.4.11) is capable of providing a complete description of the time evolution of the system and of all of its observable properties. However, there are circumstances under which the state of the system is not known in a precise manner.Under such circumstances, where the precise state of the system is unknown, the density matrix formalism can be used to describe the system in a statistical sense using the probability P(s) that the system is in the state

s. Thus in terms of P(s), we define the elements of the density matrix of the system by

$$\rho_{nm} = \sum_s P(s) C_m^{s*} C_n^s \tag{2.4.12}$$

The off-diagonal elements of the density matrix are, in certain circumstances, proportional to the induced electric dipole moment of the atom. The density matrix is useful because it can be used to calculate the expectation value of any observable quantity. Since the expectation value of an observable quantity A for a system known to be in the quantum state s is given according to Eq.(2.4.11) , the expectation value for the case in which the exact state of the system is not known is obtained by averaging Eq. (2.4.11) over all possible states of the system, to yield

$$\overline{\langle A \rangle} = \sum_s P(s) \sum_{mn} C_m^{s*} C_n^s A_{mn} \tag{2.4.13}$$

Through use of Eq. (2.4.12), this equation can alternatively be expressed as

$$\overline{\langle A \rangle} = \sum_{mn} \rho_{nm} A_{mn} \tag{2.4.14}$$

The double summation can be simplified as

$$\overline{\langle A \rangle} = \sum_n (\hat{\rho}\hat{A})_{nn} \tag{2.4.15}$$

Where $\hat{\rho}$ denotes the density operator. We have just seen that the expectation value of any observable quantity can be determined straightforwardly in terms of the density matrix. In order to determine how any expectation value evolves in time, it is thus necessary only to determine how the density matrix itself evolves in time. By direct time differentiation of Eq. (2.4.12), we finaly obtain that

$$\dot{\rho} = \frac{1}{i\hbar}[\hat{H}, \hat{\rho}]_{ij} \tag{2.4.16}$$

Equation (2.4.16) describes how the density matrix evolves in time as the result of interactions that are included in the Hamiltonian $\hat{H}$. However, as mentioned above, there are certain interactions (such as those resulting from collisions between atoms) that cannot conveniently be included in a Hamiltonian description. Such interactions can be include in the formalism by adding phenomenological damping terms to the equation of motion (2.4.16)

$$\dot{\rho} = \frac{1}{i\hbar}\left[[\hat{H}_0 - \hat{\mu}E(t), \hat{\rho}]_{ij}\right] - \Gamma_{ij}(\rho - \rho^{(0)})_{ij} \tag{2.4.17}$$

Once we obtain the time evolution of the density matrix, by using iterative procedures, perturbation expansions, variable substitution techniques and integrating with respect to time for $\rho^{(1)}_{nm}$ we get [1,12],

$$\rho^{(1)}_{nm} = \int_{-\infty}^{t} \frac{-i}{\hbar}\left[\hat{V}(t), \hat{\rho}^{(0)}\right]_{nm} e^{(i\omega_{nm}+\gamma_{nm})(t-t)}dt \tag{2.4.18}$$

and in general for $\rho^{(N)}_{nm}$

$$\rho^{(N)}_{nm} = \int_{-\infty}^{t} \frac{-i}{\hbar}\left[\hat{V}(t), \hat{\rho}^{(N-1)}\right]_{nm} e^{(i\omega_{nm}+\gamma_{nm})(t-t)}dt \tag{2.4.19}$$

Chapter 3

Materials and Methodology

3.1 Materials

For this work an intensive survey of literature from Published articles, books and thesis was assessed and carried out. Software applications; Mathlab, Latex, and Mathematica was applied for mathematical operations, graphs and editing text.

3.2 Methodology

Here in this research work we used analytic and computational methods.

3.2.1 Analytical

The analytical method that was applied for this research are Schrodinger Equation and density matrix formalism.

3.2.2 Computational

Using the analytical methods described above third-order nonlinear; absorption coefficient and refractive index changes was calculated and the obtained results are discussed and elaborated graphically.

Chapter 4

Third order Nonlinear Refractive Index and Absorption Coefficient of ZnO Quantum Dot

In this research analytical and numerical calculations have been made to describe the dependence of third order nonlinear refractive index and absorption coefficient on optical intensity and transition energy of the quantum dot. The frequency response of the real and imaginary part of third order nonlinearity is studied using compact density matrix approach. The change in the real and imaginary part of third order nonlinear susceptibility of ZnO quantum dot is investigated with respect to the change in frequency of laser source and transition energy.

4.1 Quantum mechanical treatment of third order nonlinear Susceptibility

Linear and nonlinear optical properties which are connected to intersubband transitions in semiconductor have been a subject of great interest. Due to the strong quantum confinement effects, the low-dimensional quantum systems can cause more

amplified nonlinear optical effects than bulk materials [23,26]. In practical applications, the linear and nonlinear optical properties have a wide range of potential application for high speed electro-optical modulators, for infrared photo detectors and semiconductor optical amplifiers. Moreover, the nonlinear optical properties of nanomaterials are also highly desirable for optical switching, pulse power shaping and other nonlinear optical applications. In this section we consider the third order nonlinear susceptibility in a quantum dot. Third order nonlinear optical absorption coefficient and refractive index changes is derived using the compact density matrix formalism and an iterative procedure.

4.2 The energy eigenvalues of ZnO quantum dot

A system of electrons fully confined in all three dimensions will have discrete charge and electronic states. They are often called artificial atoms or quantum dots to reflect the importance of quantization phenomena on their properties. Consider an electron in a spherical quantum dot of rigid confinement. The Schrodinger equation for such a spherical quantum dot in hard confinement is given by [20,28]:

$$\frac{-\hbar^2}{2m^*}\nabla^2\psi + \hat{V}\psi = E\psi \tag{4.2.1}$$

$$\nabla^2 = \frac{1}{r^2}\frac{\partial}{\partial r}(r^2\frac{\partial}{\partial r}) + \frac{1}{r^2 sin\theta}\frac{\partial}{\partial\theta}(sin\theta\frac{\partial}{\partial\theta}) + \frac{1}{r^2 sin^2\theta}(\frac{\partial^2}{\partial\varphi^2}) \tag{4.2.2}$$

$$V = \begin{cases} 0 & for\ r < a \\ \infty & for\ r \geq a \end{cases}$$

Due to the spherical symmetry, the wave equation of the spherical QD separated into angular and radial parts:

$$\psi(r,\theta,\varphi) = Y_{lm}(\theta,\varphi)R_{nl}(r)$$

$Y_{lm}(\theta,\varphi)$ are the spherical harmonics and $R_{nl}(r)$ are the radial wave functions [20]. The well known solution of this Schrodinger equation for rigid confinement is:

$$\psi(r,\theta,\varphi) = Y_{lm}(\theta,\varphi)\frac{1}{a}\sqrt{\frac{2}{r}}\frac{J_{l+1/2}(\beta_{nl}r)}{J_{l+3/2}(\beta_{nl}a)} \tag{4.2.3}$$

Where, a is the radius of the quantum dot, and β_{nl} *is the* n^{th} *zero of* $J_{l+1/2}(\beta_{nl}a)$. For an infinite spherical well, where V=0 for $r < a$ and is infinity otherwise, E_{nl} will be [20]

$$E_{nl} = \frac{\hbar^2\beta_{nl}^2}{2m^*a^2}$$

Here we consider the first two roots of spherical Bessel functions

i.e $\beta_{00} = \pi$ *and* $\beta_{01} = 4.49$

$$E_{00} = \frac{\hbar^2\beta_{00}^2}{2m^*a^2} = \frac{\hbar^2\pi^2}{2m^*a^2} = 0.45eV$$

$$E_{01} = \frac{\hbar^2\beta_{01}^2}{2m^*a^2} = \frac{\hbar^2(4.49)^2}{2m^*a^2} = 0.90eV$$

With the effective mass *of zinc oxide* ($m^* = 0.21m_0$), for a quantum dot of radius a=2nm

$$E_{00} = 0.45eV \text{ and } E_{01} = 0.90eV$$

For optical transition, to take place the applied photon energy must be in equal footing with $E_{01} - E_{00} = 0.45eV$

4.3 Intersubband optical Absorption coefficients and Refractive index changes

Photoabsorption process may be defined as an optical (intersubband) transition in low dimensional quantum mechanical systems[27]. The intersubband transitions in quantum wells have attracted much interest due to their unique characteristics: a large dipole moment, an ultra-fast relaxation time, and an outstanding tunability of the transition energy. The intersubband nonlinear absorption coefficients and refractive index changes depend on the transition energy. In QDs this quantity is strongly modified by intensity of a laser field [15]. In this section we consider the third order nonlinear susceptibility in a quantum dot. A brief third order nonlinear refractive index changes and absorption coefficient using the compact density matrix formalism and iterative procedures are described here. A polarized electromagnetic field interacting with the quantum-dot excites the system. The electric field vector of this electromagnetic wave is [1,19,22]

$$\tilde{E}(t) = \tilde{E}e^{-i\omega t} + \tilde{E}e^{i\omega t} = E_0 cos\omega t \tag{4.3.1}$$

Let us denote one electron density matrix as $''\rho\ ''$ for the quantum dot. Then the time evolution of the density matrix operator ρ is given by [1,10]

$$\dot{\rho} = \frac{1}{i\hbar}[\hat{H}, \hat{\rho}]_{ij} - \Gamma_{ij}(\rho - \rho^{(0)})_{ij}$$

Since $\hat{H} = \hat{H}_0 - \hat{\mu}E(t)$

$$\dot{\rho} = \frac{1}{i\hbar}\left([\hat{H}_0 - \hat{\mu}E(t), \hat{\rho}]_{ij}\right) - \Gamma_{ij}(\rho - \rho^{(0)})_{ij} \tag{4.3.2}$$

Where $\hat{H}_0$ is the Hamiltonian for the system without electromagnetic field E(t), $\rho^{(0)}$ is the unperturbed density matrix operator, and Γ_{ij} is the phenomenological damping

rate caused by the electron -phonon, electron-electron and other collision processes. Here we describe $\Gamma_{ij} = \Gamma_0 = 1/T_0 \; for \; i \neq j$. The damping constant is related to the inverse of relaxation time T_0. To solve equation (4.3.2) we have to use iterative method.

$$\rho(t) = \sum_n \rho^{(n)}(t) \tag{4.3.3}$$

with

$$\frac{\partial \rho_{ij}^{(n+1)}}{\partial t} = \frac{1}{i\hbar}\left([\hat{H}_0, \hat{\rho}^{(n+1)}]_{ij} - i\hbar\Gamma_{ij}\rho_{ij}^{(n+1)}\right) - \frac{1}{i\hbar}[\hat{\mu}, \hat{\rho}^{(n)}]_{ij}\vec{E}(t) \tag{4.3.4}$$

Considering a centrosymmetric system, a concise expression of the electronic polarization can be described as [13,16,19,21]

$$\vec{P}(t) = \epsilon_0\chi_\omega^{(1)}Ee^{i\omega t} + \epsilon_0\chi_\omega^{(3)}Ee^{3i\omega t} + c.c \tag{4.3.5}$$

Where $\epsilon_0, \; \chi_\omega^{(1)} \; and \; \chi_\omega^{(3)}$ are permittivity of free space, the linear and third order nonlinear susceptibility coefficients, respectively. For simplicity, we consider a two level system for electronic transitions. Denoting the ground state by 'g' and the exited state by 'e', we obtain [19,22]

$$\frac{d}{dt}\hat{\rho}_{eg}^{(n+1)}(t) = \left[\frac{1}{i\hbar}(E_e - E_g) - \Gamma_{ge}\right]\hat{\rho}_{eg}^{(n+1)}$$

$$-\frac{1}{i\hbar}[\hat{\rho}_{gg}^{(n)}(t) \; - \hat{\rho}_{ee}^{(n)}(t)]\mu_{eg}\vec{E}(t) - \frac{1}{i\hbar}[\mu_{ee} - \mu_{gg}]\vec{E}(t)\rho_{ge}^{(n)} \tag{4.3.6}$$

With, $\mu_{eg} = \langle e|\hat{\mu}|g\rangle \; and \; \Gamma_{eg} = \left(\frac{1}{T_g} + \frac{1}{T_e}\right)$. Equation (4.3.6) can be solved by expanding the density matrix elements as a sum of terms proportional to $e^{\pm i\omega t}$ and equating terms on both sides having the same time dependence [1,16,19,21]. Neglected the higher harmonic terms which corresponds to successive absorption or emission of

photons we considered only the steady-state response. Under this assumption the n^{th} order perturbation term $\hat{\rho}^{(n)}(t)$ can be written as :

$$\hat{\rho}^{(n)}(t) = \tilde{\rho}^{(n)}(\omega)e^{-i\omega t} + \tilde{\rho}^{(n)}(-\omega)e^{i\omega t} \tag{4.3.7}$$

Using eq.(4.3.1) and (4.3.6) and neglecting the off resonance terms we obtain the Fourier expansion coefficient [19]:

$$\rho_{eg}^{(3)}(\omega) = \left[\frac{-\tilde{E}|\tilde{E}|^2\mu_{eg}(\rho_{gg}^{(0)} - \rho_{ee}^{(0)})}{(\hbar\omega_{eg} - \hbar\omega - i\hbar\Gamma_{ge})}\right]$$

$$\left[\frac{2(\frac{1}{\Gamma_{gg}} + \frac{1}{\Gamma_{ee}})|\mu_{eg}|^2\Gamma_{ge}}{(\hbar\omega_{eg} - \hbar\omega)^2 + (\hbar\Gamma_{ge})^2} - \frac{(\mu_{ee} - \mu_{gg})^2}{(\hbar\omega_{eg} - i\hbar\Gamma_{ge})(\hbar\omega_{eg} - \hbar\omega - i\hbar\Gamma_{ge})}\right] \tag{4.3.8}$$

Where $\omega_{eg} = \frac{(E_e - E_g)}{\hbar}$, is the transition frequency.

Neglecting the first order and higher order terms,the third order electronic polarization is [23]

$$\vec{P}(t) = \epsilon_0\chi^{(3)}Ee^{(i\omega t)} \tag{4.3.9}$$

The analytical forms of the third order nonlinear susceptibility is [19,27]

$$\chi^{(3)}(\omega) = -\left[\frac{N|\mu_{eg}|^2\frac{\mu cI}{2n_r}}{\epsilon_0(\hbar\omega_{eg} - \hbar\omega - i\hbar\Gamma_{ge})}\right]$$

$$\left[\frac{4|\mu_{eg}|^2}{[(\hbar\omega_{eg} - \hbar\omega)^2 + (\hbar\Gamma_{ge})^2]} - \frac{(\mu_{ee} - \mu_{gg})^2}{(\hbar\omega_{eg} - i\hbar\Gamma_{ge})(\hbar\omega_{eg} - \hbar\omega - i\hbar\Gamma_{ge})}\right] \tag{4.3.10}$$

Where, $\mu_{eg} = \langle e|\hat{\mu}|g\rangle = \langle e|ez|g\rangle$, $\mu_{gg} = \langle g|\hat{\mu}|g\rangle = \langle g|ez|g\rangle$, μ , is the permeability of the medium, n_r refractive index of the medium, c is speed of light, N is the carrier density in this system and I is the incident optical intensity, which is given as; $I = \frac{2n_r}{\mu c}|E(\omega)|^2$

The real and imaginary parts of the third order susceptibility[] from eq.(4.3.10);

$$R_e|\chi^{(3)}(\omega)| = -\frac{\mu c}{2n_r\epsilon_0}\frac{NI|\mu_{eg}|^2}{[(\hbar\omega_{eg} - \hbar\omega)^2 + (\hbar\Gamma_{ge})^2]^2}[4(\hbar\omega_{eg} - \hbar\omega)|\mu_{eg}|^2$$

$$-\frac{(\mu_{ee}-\mu_{gg})^2}{(\hbar\omega_{eg})^2+(\hbar\Gamma_{ge})^2}\times[(\hbar\omega_{eg}-\hbar\omega)[(\hbar\omega_{eg})(\hbar\omega_{eg}-\hbar\omega)-(\hbar\Gamma_{ge})^2]-(\hbar\Gamma_{ge})^2(2\hbar\omega_{eg}-\hbar\omega)]] \tag{4.3.11}$$

$$I_m|\chi^{(3)}(\omega)| = -\frac{\mu c}{2n_r\epsilon_0}\frac{NI|\mu_{eg}|^2}{[(\hbar\omega_{eg}-\hbar\omega)^2+(\hbar\Gamma_{ge})^2]^2}[4(\hbar\Gamma_{ge})|\mu_{eg}|^2$$

$$-\frac{(\mu_{ee}-\mu_{gg})^2}{(\hbar\omega_{eg})^2+(\hbar\Gamma_{ge})^2}\times[(\hbar\Gamma_{ge})[(\hbar\Gamma_{ge})(\hbar\omega_{eg}-\hbar\omega)+\hbar^2\Gamma_{ge}\omega_{eg}]+[(\hbar\Gamma_{ge})(\hbar\omega_{eg}-\hbar\omega)-\hbar^2\Gamma_{ge}^2]]] \tag{4.3.12}$$

The real part of susceptibility is related to the change in nonlinear refractive index :

$$\frac{\Delta n(\omega)}{n_r} = \frac{1}{2n_r^2}R_e\chi(\omega) \tag{4.3.13}$$

Thus using equation (4.3.11)and (4.3.13),the change in third-order nonlinear refractive index will be [9,16,21]:

$$\frac{\Delta n^{(3)}}{n_r} = -\frac{\mu c}{4n_r^3\epsilon_0}\frac{NI|\mu_{eg}|^2}{[(\hbar\omega_{eg}-\hbar\omega)^2+(\hbar\Gamma_{ge})^2]^2}\times[4(\hbar\omega_{eg}-\hbar\omega)|\mu_{eg}|^2$$

$$-\frac{(\mu_{ee}-\mu_{gg})^2}{(\hbar\omega_{eg})^2+(\hbar\Gamma_{ge})^2}[(\hbar\omega_{eg}-\hbar\omega)[(\hbar\omega_{eg})(\hbar\omega_{eg}-\hbar\omega)-(\hbar\Gamma_{ge})^2]-(\hbar\Gamma_{ge})^2(2\hbar\omega_{eg}-\hbar\omega)]] \tag{4.3.14}$$

Third order nonlinear absorption coefficient is related with the imaginary part of $\chi^{(3)}$ as follows[16,13,27];

$$\beta^{(3)}(\omega) = \frac{\omega}{n_r}\sqrt{\frac{\mu}{\epsilon_0}}I_m\chi^{(3)}(\omega) \tag{4.3.15}$$

Comparing equ.(4.3.12) and (4.3.15) we obtain

$$\beta^{(3)}(\omega) = -\frac{\omega c}{2n_r^2}[\frac{\mu}{\epsilon_0}]^{3/2}\frac{NI|\mu_{eg}|^2}{[(\hbar\omega_{eg}-\hbar\omega)^2+(\hbar\Gamma_{ge})^2]^2}\times[4(\hbar\Gamma_{ge})|\mu_{eg}|^2-\frac{(\mu_{ee}-\mu_{gg})^2}{(\hbar\omega_{eg})^2+(\hbar\Gamma_{ge})^2}$$

$$\times[(\hbar\Gamma_{ge})[(\hbar\Gamma_{ge})(\hbar\omega_{eg}-\hbar\omega)+\hbar^2\Gamma_{ge}\omega_{eg}]+[(\hbar\Gamma_{ge})(\hbar\omega_{eg}-\hbar\omega)-\hbar^2\Gamma_{ge}^2]]] \quad (4.3.16)$$

Parameters we used in this research are $N = 1\times10^{22}/m^3$, $radius\ of\ the\ QD$ ($a_1 = 1nm, a_2 = 2nm, a_3 = 3nm$), $n_r = 3.2$, $\mu = 1.26\times10^{-6}$, $and\ \Gamma = 5\times10^{14}/s$.

The transition energies that correspond to the three different quantum radius and the relative transition dipole moments are: for $a_1 = 1nm$ ($\Delta E_1 = 1.51\times10^{-19}J,\ \mu_{eg} = -1.6\times10^{-28}C-m$), $for\ a_2 = 2nm(\Delta E_2 = 0.757\times10^{-19}J,\ \mu_{eg} = -1.7\times10^{-28}C-m)$, $for\ a_3 = 3nm(\Delta E_3 = 0.336\times10^{-19}J,\ \mu_{eg} = -1.8\times10^{-28}C-m)$.

4.4 graphical descriptions.

The variation of the change in third order nonlinear refractive index with respect to three different transition energies is described in fig.4.1

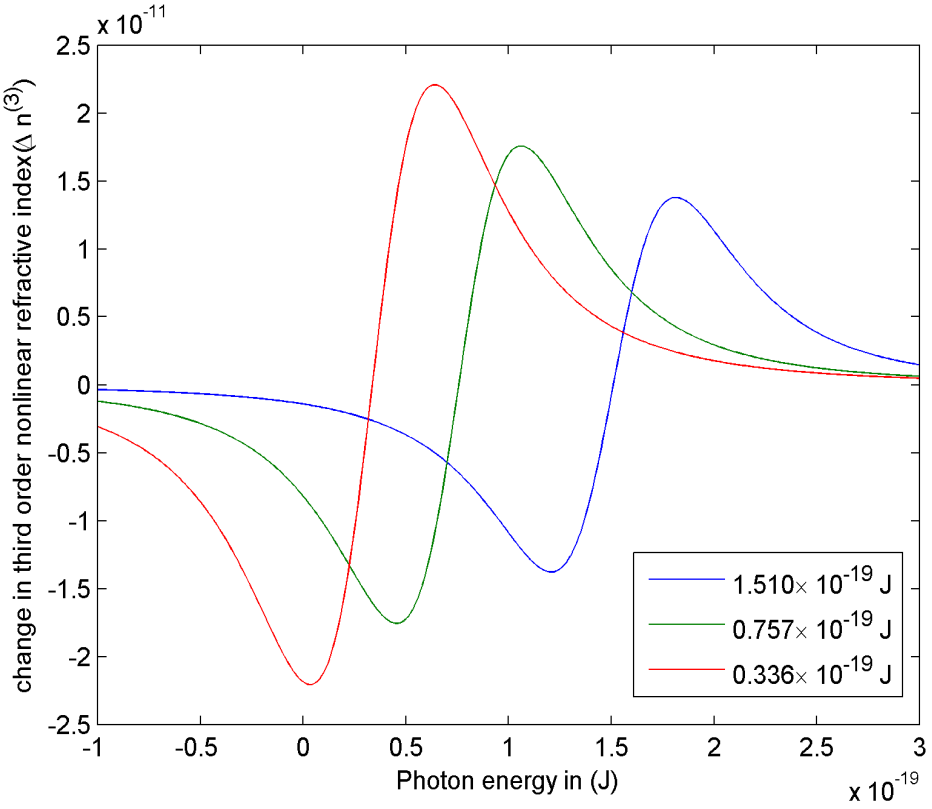

Figure 4.1: *The variation of the change in third order nonlinear refractive index versus transition energy for* $I = 7.189 \times 10^8 W/m^2$.

As it is observed from figure 4.1 there is a blue shift depending on an increment of transition energy from $0.336 \times 10^{-19} J$ *to* $1.5 \times 10^{-19} J$.

Moreover, the variation of change in third order refractive index is studied as a function of optical laser intensity. Fig.4.2 shows the variation of change in third order refractive index versus photon energy for thre different optical intensities $I = 1 \times 10^{10} W/m^2$, $5 \times 10^{10} W/m^2$ *and* $10 \times 10^{10} W/m^2$ are described in figure 4.2. The result

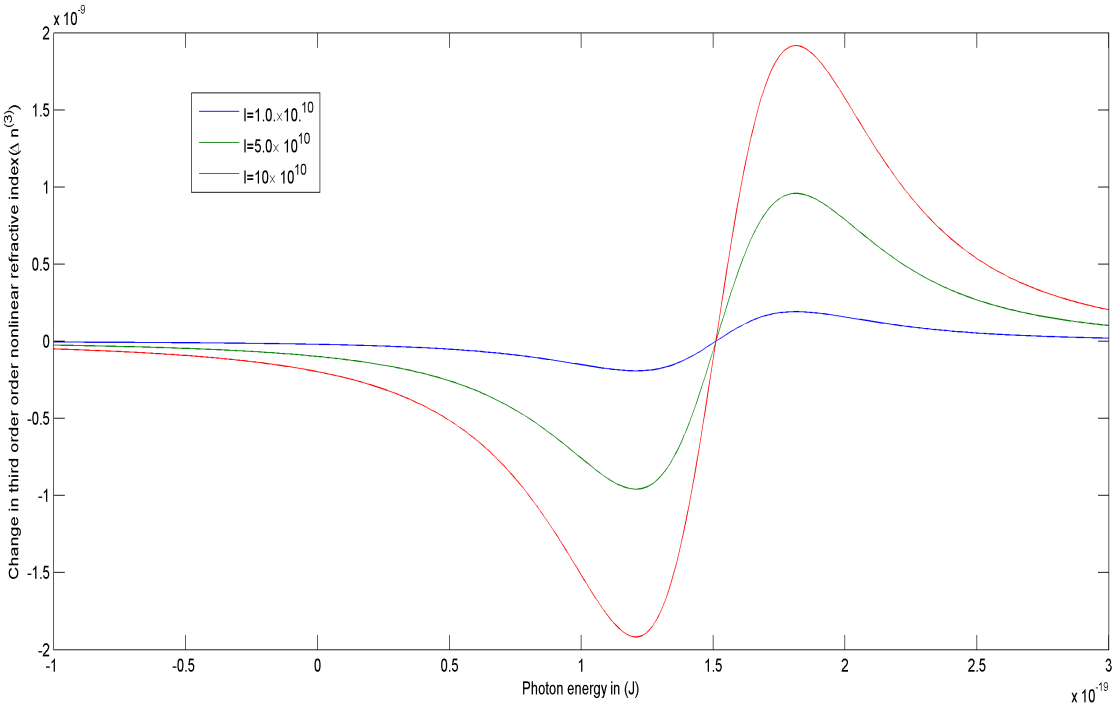

Figure 4.2: *The change in third order nonlinear refractive index versus photon energy. For* $I = 1 \times 10^{10} W/m^2$, $5 \times 10^{10} W/m^2$ *and* $10 \times 10^{10} W/m^2$

clearly shows that the magnitude of the change in third order nonlinear refractive index increases as the intensity of the laser source increases. That is high optical intensity induces large nonlinearity.

The third order nonlinear absorption coefficient is also studied as a function of the three different transition energy($1.5 \times 10^{-19}J, 0.75 \times 10^{-19}J,\ 0.336 \times 10^{-19}J$), which was graphically demonstrated in figure 4.3. As it is observed from the figure, there

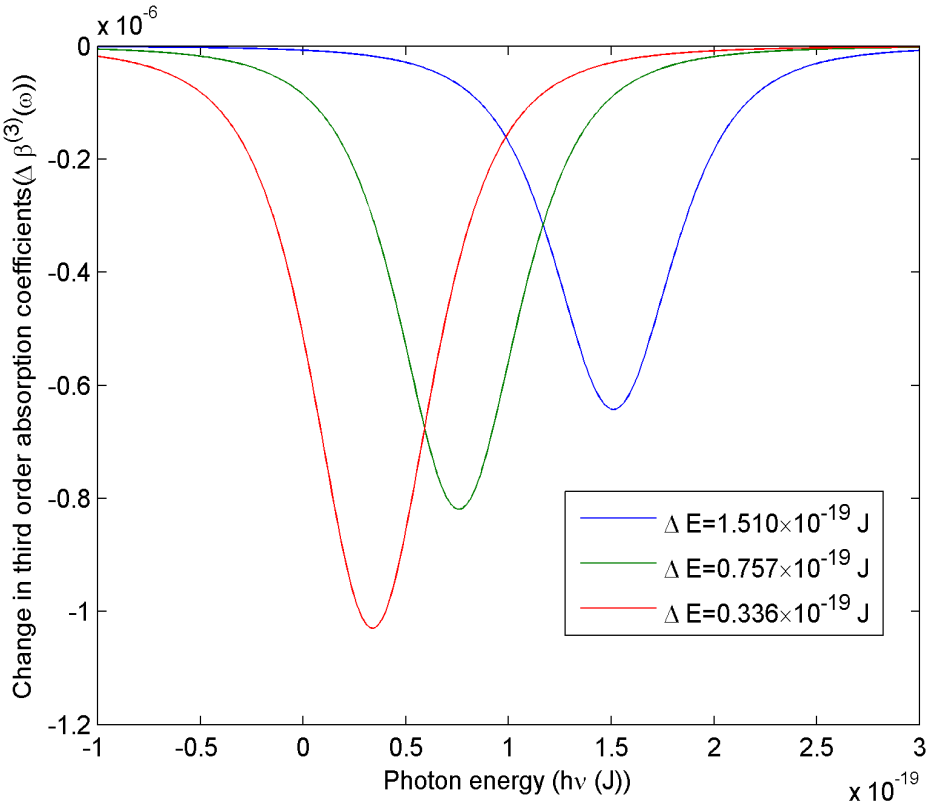

Figure 4.3: *The change in third order nonlinear absorption coefficient versus photon energy, for* $I = 7.189 \times 10^8 W/m^2$.

is a blue shift as the value of transition energy increases. That is as the transition energy increases, the spectra shifts towards the higher energy. However the absolute magnitude of the absorption coefficient decreases.

Finally, we have studied the effect optical Intensity on third order nonlinear absorption coefficient. The graphical analysis is made for the intensities $I = 1 \times 10^{10} W/m^2, 5 \times 10^{10} W/m^2$ *and* $10 \times 10^{10} W/m^2$, respectively. As it is seen from figure

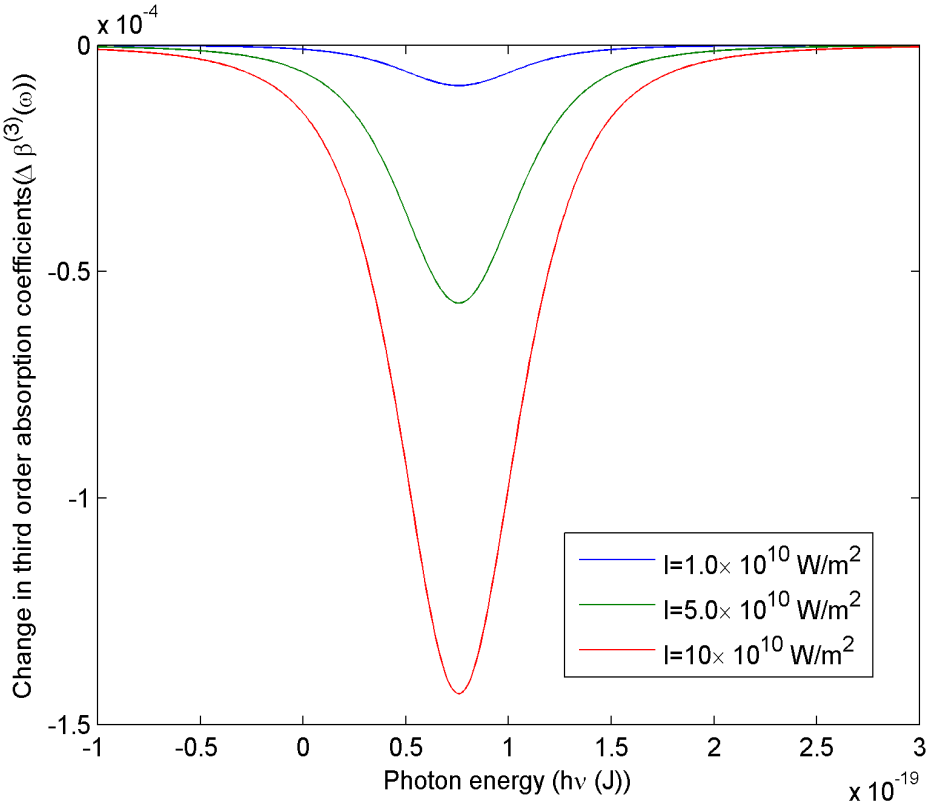

Figure 4.4: *The change in third order nonlinear absorption coefficient versus photon energy for three different values of optical intensity.*

4.4, the third order nonlinear absorption coefficient increases with an increment of the applied optical intensity of the electromagnetic field radiation.

Chapter 5

Conclusion

In this research work we have performed a theoretical study of third order nonlinear absorption coefficient and refractive index changes of spherical QD as a function of transition energy and laser source intensity. We developed third order NL absorption coefficient and refractive index changes using density matrix formalism. The obtained result show that as the transition energy increases from $0.336 \times 10^{-19} J$ *to* $1.5 \times 10^{-19} J$ a blue shift was observed in both absorption coefficient and refractive index changes. Moreover as the intensity of the laser source increases from $1 \times 10^{10} W/m^2$ *to* $10 \times 10^{10} W/m^2$ an amplified magnitude of third order absorption coefficient and refractive index changes resulted, that indicates high laser intensity induces strong third order nonlinear effects, which agreed with [13]. Theoretical study can make a significant contribution to experimental studies and we hope that this research work can have a significant input.

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Graduate Program: **MSc.in Physics**

1. Course Work Performance

Course Code	Course Title	Cr. hr	Number Grade	Rank **	Remark
Phys699	**MSc. Thesis**	**6**	**85.45**	**Excellent**	

** Excellent, Very Good, Good, Satisfactory, Fail.

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Third order Nonlinear Refractive Index and Absorption coefficient of Zinc Oxide (ZnO)Quantum Dot

2. Board of Examiners decision Mark × in one of the boxes. Pass [×] Failed [ ]

   If failed, give reasons and indicate plans for re-examination.

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DECLARATION

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October, 2017