
# NONLINEAR OPTICAL PROPERTIES OF CYLINDRICAL QUANTUM WIRES OF ZnO WITHIN APPLIED ELECTRIC FIELD USING PARABOLIC CONFINING POTENTIAL

By

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DECLARATION

I hereby declare that this Masters thesis is my original work and has not been presented for a degree in any other university, and that all sources of material used for the proposal have been duly acknowledged.

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# Abstract

One of the most intensively explored classes of semiconductor structures is the class of quantum wires. With the advances in semiconductor growth techniques it is possible to fabricate quantum wires of different sizes and geometries from different materials. Cylindrical quantum wire made of ZnO is one of such geometries. This cylindrical structure can be described using a parabolic potential model. In this study the energy eigenvalue of restricted electrons was calculated for ZnO cylindrical quantum wire. Using this energy eigenvalue the nonlinear optical properties such as index of refraction and absorption coefficient were investigated with the help of density matrix formalism. From the result of this study the change in index of refraction $\Delta n(\omega)$ and absorption coefficient $\Delta\beta(\omega)$ were calculated numerically and represented graphically for the applied electric field. It is observed that the nonlinear refractive index and absorption coefficient are depend on the optical intensity and their magnitude is amplified as the optical intensity increases.

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# Contents

# List of Figures

# List of Tables

# 1

# Introduction

## 1.1 Background of the study

Nanotechnology is an essentially modern scientific field that is constantly evolving as commercial and academic interest continues to increase and as new research is presented to the scientific community[1]. The field's simplest roots can be traced, albeit arguably, to 1959 but its primary development occurred in both the eighties and the early nineties[2,3]. It owes its existence to the astonishing development within the field of micro electronics[4]. Since the invention of the first semiconductor transistor in 1947 by the scientists of Bell Labs, the semiconductor industry has grown at an incredible pace, fabricating faster, smaller, more powerful devices while manufacturing in larger volume at lower costs[5,6]. The integrated circuit invented in 1958, has resulted an exponential growth in the number of transistors per microchip and an associated decrease in the smallest width of the wires in the electronic circuits.

The investigation of semiconductor low-dimensional structures[7] has a key role on the development of nanotechnology and future nanoscale devices due to their unique electrical and optical properties[8]. These materials often exhibit remark-

able mechanical as well as electrical, optical, and electromagnetic properties that are quite different from those of the corresponding bulk materials[9,10]. As a result, there exists a possibility of a continuous tuning of the optical and electronic properties by varying the size of the particles[11]. Recent advances in growth techniques made possible the fabrication of one dimensional structure quantum wires [12] with nanoscale diameters such as nanowires, nanorods, and nanotubes have attracted considerable attention due to their peculiar structure characteristics and size effects[13], where electrons and holes are confined in two dimensions, with only one free dimension[14,15]. Quantum wires based on radial semiconductor heterostructures [16] have been successfully used in opto-electronic systems and biological sensors, which require a strong confinement of the charge carriers[17]. That is why such materials can be viewed as promising candidates for future applications in the field of catalysis, sensor technology, transistor, electrode materials, logic circuits, and laser working[18]. It is necessary to note that ZnO shows quantum confinement effects in the experimentally accessible range of sizes less than 7 nm which is a technologically important material with widespread use and it has been of much interest in synthesizing ZnO nanocrystals of various sizes in the past few years[19].

## 1.2 Statement of the problem

In this thesis, we want to know the property of low dimensional semiconductor systems in comparison with their bulk system. Quantum confinement of charge carriers causes quantized energy in low dimensional system. The value of these ener-

gies will be calculated by using Schrodinger equation. The ground and first excited states of cylindrical quantum wire is already studied. The focus of this work is the effect of applied electric field to the cylindrical quantum wire ZnO.

Recent advances in nanotechnology have made it possible to fabricate nanowires of different sizes and geometries[20]. In nanostructures cylindrical surfaces show physical interesting properties. These structures can be described using a parabolic potential model and it is one of the types confining potentials. The parabolic confining potential is also introduced for technological reasons. With advances in material growth techniques, such as atomic layer epitaxy, etc.[21] the growth of single atomic layers of good quality has become possible, which has allowed potential profiles with reasonable shapes such as parabolic shapes and stepped shapes, etc[22]. In this study we have determined the optical properties of ZnO cylindrical quantum wire in parabolic confining potential using density matrix formalism[23]. The main purpose of this study is calculating the energy eigenvalue of ZnO cylindrical quantum wire for applied electric field, and determining its nonlinear optical properties.

**Research questions:**

✓ What is the confining potential we use to describe cylindrical quantum wire?

✓ How can we calculate energy eigenvalue of confined electrons?

✓ What are the linear and nonlinear optical properties of ZnO cylindrical quantum wire?

✓ What are the applications of nonlinear optical properties of ZnO cylindrical quantum wire?

## 1.3 Objectives of the study

### 1.3.1 General objectives

The general objective of this study is to determine the energy eigen values of ZnO cylindrical quantum wires and to describe its optical properties of ZnO cylindrical quantum wire with applied electric field.

### 1.3.2 Specific objectives

The specific objectives of this study are:

✓ To determine linear and nonlinear refractive indices of ZnO cylindrical quantum wire for applied electric field.

✓ To determine linear and nonlinear absorption coefficients of ZnO cylindrical quantum wire for electric field applied.

## 1.4 Significance of the study

Optical properties of low-dimensional semiconductor structures are important part of the modern physics of semiconductors. A large number of theoretical and experimental works have been devoted to the study of optical properties in low dimensional semiconductor nanostructures. It is to be noted that the study of optical properties of low dimensional semiconductor structures is important, not only to know, but also in the fabrication and subsequent working of electronic and opti-

cal devices based on such systems. Also this study helps us to know the energy eigenvalue and nonlinear optical properties of ZnO cylindrical quantum wire when electric field is applied to it.

## 1.5 Scope of the Study

The research is intended to evaluate the index of refraction and absorption coefficient among the nonlinear optical properties of cylindrical quantum wires of ZnO for the applied electric field by density matrix formalism.

## 1.6 Limitation of the Study

Due to time constraint, the scope of this study was limited only to determine the linear and nonlinear index of refraction and absorption coefficients among many other optical properties of ZnO cylindrical quantum wire for confined electrons in parabolic confining potential using density matrix formalism in applied electric field.

## 1.7 Outlines of the thesis

This study was organized as follows: In chapter 1, we have discussed some background concepts of low dimensional semiconductor systems including the statement of the problem, objectives and significance of the study. In chapter 2, we review the theoretical concept of low dimensional systems, calculate their density of states and we derive the general expression of wave function and energy eigenvalue of single electron for cylindrical quantum wire with parabolic confinement in ap-

plied electric field. In chapter 4, we introduce the analytical and numerical methods used to carry out the study and finally in chapter 5, we draw some conclusions.

# 2

# Review of Related Literature

## 2.1 Introduction

Civilization has always been modified by man's relationship with materials. Currently materials have become such an integral part of our society, that they are often under appreciated or even overlooked. But much of the technological progress is directly or indirectly dependant on the availability of advanced materials with improved functions and desirable properties. It is often forgotten, that there is enormous science behind these investigations and developments. The definition of materials as 'substances having properties which make them useful in machinery, structures, devices and products clearly connects the materials with function. The nanoscientist's future goal shall be to acquire expertise and knowledge to enable us to build and organize nanoscale objects of one cluster of atoms or even one atom at a time instead of manipulating the bulk, thus providing a reproducible method of preparing materials that are perfect in size and shape. At present, nanoscience is concerned with the development of novel methods for the synthesis and characterization of thin films within in the size range of about 1nm to 100nm. The interest in nanoscale objects is due to the exhibition of novel electronic, optical, transport,

photochemical, catalytic and magnetic behavior depending on composition, size and shape of the particles. When we are able to gain control over the size and shape of the particles, further enhancement of material properties and device functions will surely be possible. Each change in the composition or size of the materials can also lead to different physical and chemical properties, thus providing an array of new applications[24]. When the size of the material is in the nanometer range, the many usual insignificant properties of the material such as quantum mechanical properties and thermodynamic properties become dominant and cause special properties to nanomaterials. Thus nanotechnology is the creation and use of materials, devices and systems that exploit the novel properties arising from the structure and function of the matter and are composed of materials in size of nanometer range[25]. Materials at the nanoscale are attracting more attention due to their fascinating properties making them with improved properties compared to their bulk counterparts due to large surface area and quantum confinement. They are expected to play an important role in diagnostics, increasing the efficiency of energy production, development of novel semiconductor devices, optoelectronic devices, displays and quantum computers[26].

Even long before the start of ”nanoera”, people were coming across various nano-sized objects and the related nanolevel processes, and using them in practice. However,intuitive nanotechnology antiquities developed spontaneously,without due understanding of the nature of these objects and processes.For example, the fact that small particles of various substances possessed properties different to those of the same substances with larger particle size was known for a long time, but the rea-

son for this was not clear. Thus, people were engaged in nanotechnology subconsciously, without guessing that they were dealing with the nanoworld phenomena. In many instances secrets of ancient nanoproduction simply passed from generation to generation, without getting into the reasons why the received materials and products derived from them acquired their unique properties.

Thousands of years B.C people knew and used natural fabrics:flax, cotton, wool, silk. They were able to cultivate them and process into products. What makes these fabrics special is the fact that they have a developed network of pores with the size of 1-20 nanometers i.e., they are typical nanoporous materials. Due to their nanoporous structure natural fabrics possess high utilitarian properties: they absorb sweat well, quickly swell and dry. Since ancient times people mastered the ways of making bread, wine, beer, cheese and other foodstuffs, where the fermentation processes on nanolevel are critical.

The British museum boasts Licurg's bowl as part of its heritage - an outstanding product of glass makers of Ancient Rome. This bowl possesses unusual optical properties: it changes color with change of location (inside or outside) of the light source. In natural light the bowl is green, if illuminated from within, it turns red. The analysis of fragments of the bowl, carried out in the laboratories of General Electric in 1959 for the first time, showed that the bowl consists of usual soda-lime-quartz glass and has about 1% of gold and silver, and also 0.5% of manganese as components. The researchers then assumed that the unusual color and disseminating effect of glass is provided by colloidal gold. Later, when research techniques became more advanced, scientists discovered particles of gold and silver from 50 to

100 nanometers in size using an electronic microscope and roentgenograms. It is these particles which are responsible for the unusual coloring of the bowl[27].

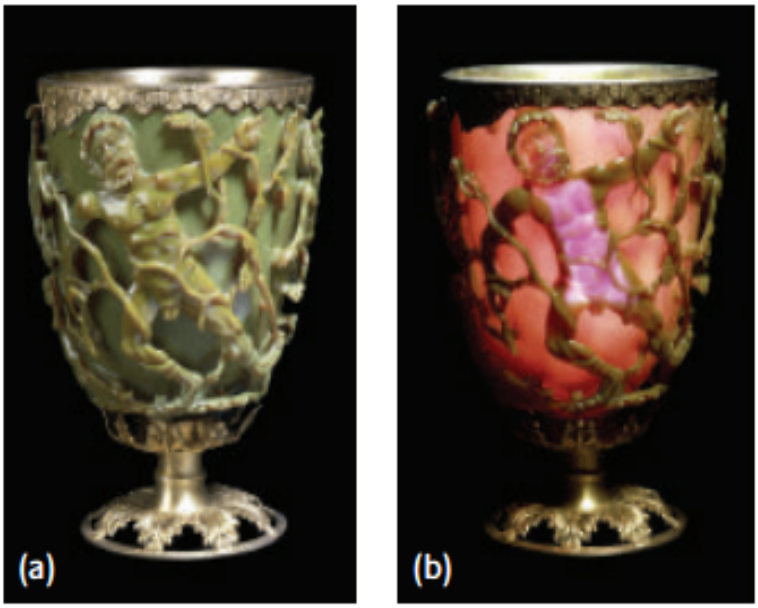


Figure 2.1: The Licurg's Cup in reflected (a) and transmitted (b) light.

Though the very first semiconductor transistor was made from germanium (Ge), silicon (Si) became the semiconductor of alternate choice as a result of the low melting point of Ge that limits high temperature processes and the lack of a naturally occurring germanium oxide to prevent the surface from electrical leakage. Gallium arsenide (GaAs) is a direct bandgap semiconductor, whereas Si is indirect, making GaAs better suited for optoelectronic devices. However, physical properties required for high power, high temperature electronics and UV/blue light emitter applications are beyond the limits of Si and GaAs. It is essential to investigate alternate materials and their growth and processing techniques in order to achieve such

devices. Wide bandgap semiconductors exhibit inherent properties such as larger bandgap, higher electron mobility and higher breakdown field strength. Therefore, they are suitable for high power, high temperature electronic devices and short wavelength optoelectronics.

From the early 1980s until just a few years ago, gallium arsenide (GaAs) field effect transistors (FETs) were the mainstay of the solid state high power amplifiers (HPA) industry for radio frequency (RF) transmission. Invented more than 30 years ago, the GaAs FET quickly gained acceptance over siliconbased devices due to its superior efficiency and ability to operate at much higher frequencies. In similar fashion, gallium nitride (GaN) FETs are now rapidly supplanting GaAs FETs for almost entirely the same reasons: better efficiency and the ability to operate at even higher frequencies. GaN FETs also outperform GaAs FETs in other key metrics, such as maximum temperature, gain, power handling, size and weight. The adoption of GaN devices is occurring at a rapid pace, with foundries around the world racing to create new GaN devices[28].

ZnO is an attractive material for applications in electronics, photonics, acoustics, and sensing[29]. In optical emitters, its high‘ exciton binding energy (60 meV) gives ZnO an edge over other semiconductors such as GaN if reproducible and reliable p-type doping in ZnO were to be achieved, which currently remains to be the main obstacle for realization of bipolar devices[30]. On the electronic side, ZnO holds some potential in transparent thin film transistors (TFTs) owing to its high optical transmittivity and high conductivity. A significant part of the recent research in the field of ZnO-based devices and applications deals with ZnO nanostructures

(nanowires, nanobelts, etc.) and their integration with the mainstream semiconductor materials such as Si, GaN, and organic semiconductors[31].

Low dimensional systems such as semiconductor quantum dots and quantum wires have fascinating and technologically useful optical and electric properties. Studies on these systems advance our knowledge on low dimensional physics and chemistry. Semiconductor quantum wires exhibit novel electric and optical properties owing to their unique structural one-dimensionality and possible quantum confinement effects in two dimensions. Quantum wires have been evaluated for potential applications as laser,light-emitting diodes, and photodetectors[32].

Semiconductor quantum wires (QWRs) has been studied intensively worldwide for a wide spectrum of materials. Such one-dimensional (1D) nanostructures are not only interesting for fundamental research due to their unique structural and physical properties relative to their bulk counterparts, but also offer fascinating potential for future technological applications. QWRs are promising candidates for optoelectronic and microelectronic devices, following the next generation of classical heterostructures and QWs which are now widely used in many applications, as for instance, heterostructure-based light-emitting diodes, heterostructure bipolar transistors, high-electron-mobility transistors and double heterostructure laser. In particular, a QWR structure has been proposed as a 1D active region for a semiconductor laser where the electron and hole carriers are allowed to move only in one direction[33].

Low-dimensional semiconductor structures are of considerable importance in the modern electronics and optoelectronics industries. This has led to the develop-

ment of crystal growth techniques which now routinely make semiconductor layers with atomic precision on the layer thickness. The application of low-dimensional structures in quantum optics comes as a spin-off from this technological progress.

**Quantum confinement**

Electron waves are characterized by their de Broglie wavelength $\lambda_{deB}$ defined by:

$$\lambda_{deB} = \frac{h}{p}, \tag{2.1}$$

where p is the linear momentum. The electrons in the conduction band of a semiconductor are free to move in all three directions, and their de Broglie wavelength is governed by the thermal kinetic energy at temperature T:

$$E_{thermal} = \frac{p_i^2}{2m_e^*} \sim \frac{1}{2}k_B T, \tag{2.2}$$

where $m_e^*$ is the effective mass and the subscript i refers to one of the Cartesian axes x, y, or z. This gives a de Broglie wavelength of order:

$$\lambda_{deB} \sim \frac{h}{\sqrt{m_e^* k_B T}} \tag{2.3}$$

In normal circumstances, the de Broglie wavelength is much smaller than the dimensions of the crystal, and the motion is governed by the laws of classical physics. However, when one or more of the dimensions of the crystal is comparable to $\lambda_{deB}$, then the motion in that direction will be quantized. This phenomenon is called quantum confinement.

It is apparent from eqn (2.3) that the length scale for the transition from classical to quantum behaviour depends on both the temperature and the effective mass. In a typical semiconductor with $m_e^* \sim 0.1m_0$, we require length scales of about 10nm or

less to observe quantum confinement effects at room temperature.

There are three general classifications of quantum confinement effects. If the motion is confined in one direction (e.g. the z-direction), the structure is called a quantum well. The electrons are free to move in the other two directions (i.e. the x- and y-directions) and so we have free motion in two dimensions and quantized motion in the third. If the motion is confined in two directions the structure is called a quantum wire. This has free motion in one dimension (e.g. the x-axis) and quantized motion in the other two directions. Finally, if the motion is confined in all three directions the structure is called a quantum dot, or alternatively a quantum box. The motion of the electrons in a quantum dot is quantized in all three directions. The general scheme of classifying quantum-confined structures is illustrated schematically in Fig.(2.2), and summarized in Table (2.1).

| structure | Quantum confinement | Free motion |
|---|---|---|
| Bulk | None | x,y,z |
| Quantum well | 1-D | x,y |
| Quantum wire | 2-D | x |
| Quantum dot | 3-D | None |

Table 2.1: General scheme of quantum confinement.

The main effect of the quantum confinement is to modify the energy spectrum and the density of states. The electrons in a bulk semiconductor can have any energy above the band-gap energy $E_g$ and the density of states is proportional to $(E-E_g)^{1/2}$. This is a consequence of the free motion in all three dimensions. The effective di-

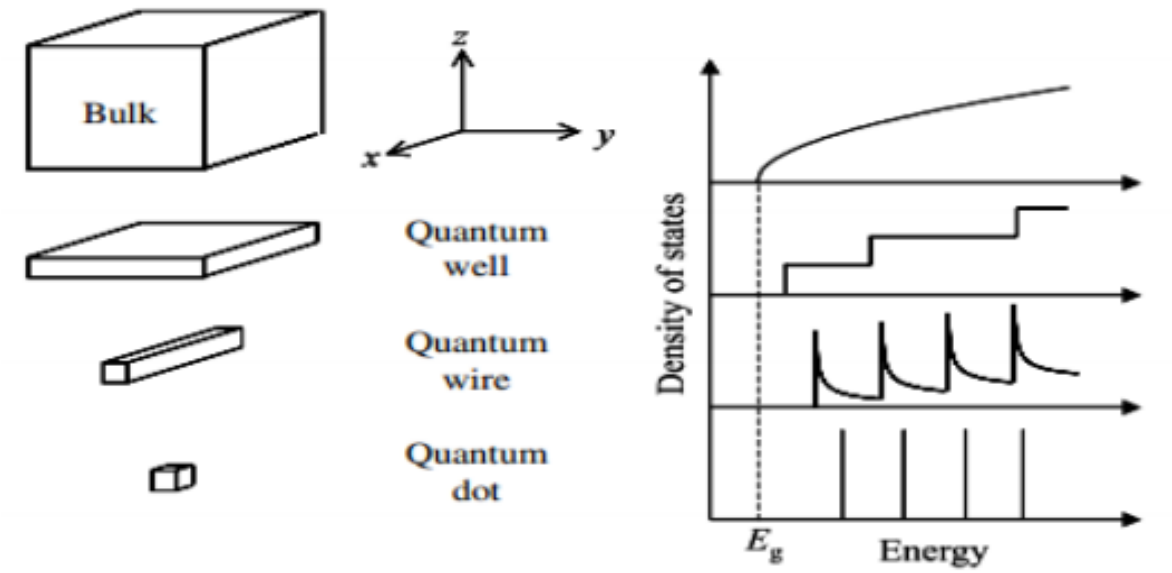


Figure 2.2: Schematic representation of quantum wells, wires, and dots.The generic shape of the density of states function for electrons in the conduction band of a semiconductor with band gap $E_g$ is shown for each type of structure.

mensionality of the system decreases as the electrons are confined in each new direction, which alters the functional form of the density of states and increases the effective band gap.[34]

Just as there is a change in the density of states moving from the bulk (3D) crystal to a quantum well (2D), there is a further change in the density of states on moving to quantum wires (1D) and quantum dots (0D).[35]

## 2.2 Optical Properties of ZnO quantum wires

Since 1970s Scientific research into electronic structure had been devoted to two dimensional structure quantum wells, the new and unusual properties of quasi-two-dimensional systems, which promise applications mostly in electronics and

opto-electronics, have attracted the attention of many researchers. This in turn has resulted in a rapid development of production technology and extensive research. This rapid progress in technology made it possible to create the quantum wires[36]. Nano-wires are new materials, which have characteristics with sometimes extraordinary mechanical, electrical, thermal and multifunctional properties very different from bulk properties[37]. By creating nanostructures; it is possible to control the fundamental properties of materials without change of chemical composition. The attractive world of low dimensional systems, together with the fabrication of functional nanostructured arrays will play a major role in the new trend of chemical and biochemical nanotechnology. Nano-wires can be used for tunable transport of electrons with electronic properties strongly influenced by little perturbations on the surface, for giant surface-to-volume ratio enhancements which are important for chemical/biochemical applications, and for generation of well defined molecular patterns on bio-sensor surfaces[38]. The optical properties of quantum wires can undergo significant changes when an electric field is applied perpendicular to the wires. This effect is called the quantum confined Stark effect, and it has been extensively studied in the past few years[39]. In the bulk semiconductor material (several times bigger than 10 nm), charge carriers can have a range of energies. These energies are so close together that they can be described as continuous. There is a certain forbidden range of energies called band gap. Almost all carriers naturally occupy the energy levels below the band gap (valence band) and only very few of them are in the conduction band (above the band gap). They can jump to the conduction band when they get additional energy from outside (heat, radiation, etc.)

and they leave a hole in the valence band. This electron-hole pair is called an exciton which is the true nature of a charged dot and has a lot of properties similar to the hydrogen atom.[40]

Zinc oxide (ZnO) is a II-VI semiconductor with a hexagonal wurtzite crystal structure, like GaN and AlN. Owing to its wide (Eg = 3.37 eV) direct band-gap, it offers great potential for the fabrication of optoelectronic devices. Recent years have witnessed increased interest in this transparent oxide for a wide range of applications due to its interesting properties. When doped with aluminium, it shows low electrical resistance combined with high optical transmittance in the visible and near infrared range, thus being competitive with indium tin oxide for application as a transparent electrode[41] One attractive fabrication method of such quantum wires has recently been reported; it is based on selective area growth using reduced pressure organometallic vapor phase epitaxy[42]. The Blue-laser diodes are a kind of quantum dot lasers which succeeds in minimizing temperature sensitive output fluctuations, something that not possible with previous semiconductor lasers. The blue-lasers diodes are made of GaN and used in optical data communications and optical networks. The commonly seen commercial product of blue-laser diodes is used as light source of High Definition DVD. A quantum wire application is nanobarcodes which is used in medical field. Nanobarcodes are made of different quantum wires of different metals that have different reflectivity. Barcode readout is accomplished by bright field reflectance imaging, typically using blue illumination to enhance contrast between Au and Ag stripes. There are 2 ways to realize nanodevices. One of them is based on the current integrated circuits to minimize the line

width. It is called top-down approach. The electronic devices only shrink in size and the basic structure of electronic devices do not change. The other way is called bottom-up approach. It is totally different from the structure of current integrated circuit and it uses quantum dot, quantum wire, and quantum well to create nano devices such as single electron transistor and single electron memory[43]. By measuring the photocurrent, one can get information about absorption effects in single nanowires[44] During the early 1980s, the so-called quantum size effects (QSE), characterized by a blue-shift of their optical spectra, has been observed in a large range of strongly confined systems. It comes from a widening of the semiconductor optical band gap, due to the increase of the charge carriers confinement energy. [45]

## 2.3 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. The optical properties of semiconductors at low light levels are often referred to as linear properties in contrast to the nonlinear optical properties. 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 (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 $\tilde{P}(t)$, of a material system depends on the strength $\tilde{E}(t)$ of an applied optical field. In the case of conventional (i.e., linear) optics, the induced polarization depends linearly on the electric field strength in a manner that can often be described by the relationship

$$\tilde{P}(t) = \varepsilon_0 \chi^{(1)} \tilde{E}(t) \tag{2.4}$$

where the constant of proportionality $\chi^{(1)}$is known as the linear susceptibility and $\epsilon_0$ is the permitivity of free space. In nonlinear optics, the optical response can often be described by generalizing Eq (2.4) by expressing the polarization $\tilde{P}(t)$ as a power series in the field strength $\tilde{E}(t)$ as

$$\tilde{P}(t) = \varepsilon_0[\chi^{(1)}\tilde{E}(t) + \chi^{(2)}\tilde{E}(t) + \chi^{(3)}\tilde{E}(t) + ...] \tag{2.5}$$

$$= \tilde{P}^1(t) + \tilde{P}^{(2)}(t) + \tilde{P}^{(3)}(t) + ... \tag{2.6}$$

The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second- and third-order nonlinear optical susceptibility, respectively. For simplicity,we have taken the fields $\tilde{P}(t)$ and $\tilde{E}(t)$ to be scalar quantities in writing Eqs.(2.4) and (2.5). We treat the vector nature of fields; in such a case $\chi^{(1)}$ becomes a second-order tensor, $\chi^2$, becomes a third-rank

tensor, and so on.

We shall refer to $\tilde{P}^{(2)}(t) = \epsilon_0 \chi^{(2)} \tilde{E}_2(t)$ as the second-order nonlinear polarization and to $\tilde{P}^3(t) = \epsilon_0 \chi^{(3)} \tilde{E}_3(t)$ as the third-order nonlinear polarization [46].

Any real, physical oscillating system will exhibit a nonlinear response when it is overdriven. In an optical system, a nonlinear response can occur when there is sufficiently intense illumination. Third order optical nonlinearities cover a vast and diverse area in nonlinear optics[47]. Compared to the linear optical properties, there is less understanding of the nonlinear optical properties of ZnO [48]. The linear and nonlinear optical properties of semiconductors are the subject of much current theoretical and experimental interest.Among the various nonlinear optical (NLO) materials investigated, wide bandgap semiconductors, especially zinc oxide (ZnO), have attractive nonlinear properties that make them ideal candidates for NLO based devices. ZnO is a wide and direct bandgap II-VI semiconductor with a bandgap of 3.37 eV and a high exciton binding energy of 60 meV and having many applications, such as solar cell, luminescent material, heterojunction laser diode, and UV laser. The optical properties of this material are currently the subject of tremendous investigations, in response to the industrial demand for optoelectronic devices that could operate at short wavelengths. Also, there is a significant demand for high nonlinear optical materials, which can be integrated into an optoelectronic device with a relatively small limiting threshold. The field of nanocomposite materials has been widely recognized as one of the most promising and rapidly emerging research areas because of their enhanced luminescence and fast nonlinear response that can be utilized in making them as potential optical devices. Significant

investigations have been done in the photophysical and photochemical behavior of single and multicomponent metal, semiconductor and dielectric nanoclusters. Such composite materials are especially of interest in developing efficient light-energy conversion systems and optical and microelectronic devices. Recent investigations have shown that ceramic composites having nanosized metal particulate dispersions show excellent optical, electrical, and mechanical properties[47]. In recent years, several publications considered one dimensional ZnO nanostructures such as nanorods, nanowires, nanobelts, nanotubes, and nanorings etc. These materials have attracted great interest due to their promising applications in electronic, optoelectronic and sensing devices. In particular, ZnO nanorods present excellent crystalline quality, high surface to volume ratio and high electron mobility. Among the huge variety of ZnO nanostructures, nanorods and nanowires(NWs) have undoubtedly been the focus of most studies since their geometries allow the preparation of arrays of well controlled uniformity and use as building blocks of many nanoscale devices. As well as for ZnO thinfilms, second harmonic generation (SHG) from ZnO NWs grown by different methods has been widely investigated. Studying SHG in ZnO NWs started few years ago and lot of information/knowledge has been gathered over the years by the nanoscale research community. Previous studies reported efficient generation of SHG signal from various types of ZnO nanostructures, including NWs. Therefore, with improvement in the fabrication technique for large area ZnO NWs, it can be widely used as one of the best SHG materials. It has been discovered that the magnitude of SHG is strongly influenced by crystal orientation, aspect ratio, crystal symmetry modification, and so on. There-

fore, by controlling such parameters one can tune the SHG of ZnO NWs. However, effective implementation of the knowledge gathered over the past years to ZnO to get the best SHG performance is yet to be achieved. Strategies for raising the optical nonlinearities of materials are an active research theme with rich and broad implications/applications[49].

In semiconductors at low temperatures, there are few (intrinsic) electrons in the conduction band. Consequently, if the incident light has a frequency lower than the bandgap frequency ($\nu_G = \frac{\xi_G}{h}$), there is no optical absorption. When the optical frequency is above the bandgap frequency, electronic interband transitions are possible and absorption is high. Thus, semiconductors act as low pass filters. They pass light below the bandgap frequency and strongly absorb light above the bandgap frequency. 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. [50]

**In nonlinear optics**(When E is very high):

✓ The refractive index, and consequently the speed of light in an optical medium, does change with the light intensity.

✓ The principle of superposition is violated.

✓ Light can alter its frequency as it passes through a nonlinear optical material(e.g., from red to blue).

✓ Light can control light; photons do interact.

The optical properties of materials are described through the real and imaginary parts of the dielectric constant. 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.[51]

## 2.4 Density Matrix formalism

We calculate the nonlinear optical susceptibility through use of the density matrix formulation of quantum mechanics. We use this formalism because it is capable of treating effects, such as collision 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 laws of quantum mechanics. If a quantum-mechanical system (such as an atom) is known to be in a particular quantum-mechanical state that we designate S, we can describe all of the physical properties of the system in terms of the wave function

appropriate to this state. The time evolution of the operator $\varrho$ can be predicted directly from the schrödinger equation.Since $\varrho(t)$ is given by:

$$\varrho(t) = \sum_{\alpha=1}^{z} \left|\Psi^{(\alpha)}(t)\right\rangle\left\langle\Psi^{(\alpha)}(t)\right| \tag{2.7}$$

$$\frac{\partial\varrho}{\partial t} = \sum_{\alpha=1}^{Z}\left[\left(\frac{\partial}{\partial t}\left|\Psi^{(\alpha)}(t)\right\rangle\right)\left\langle\Psi^{(\alpha)}(t)\right| + \left|\Psi^{(\alpha)}(t)\right\rangle\left(\frac{\partial}{\partial t}\left\langle\Psi^{(\alpha)}(t)\right|\right)\right] \tag{2.8}$$

$$= \frac{1}{i\hbar}\sum_{\alpha=1}^{z}\left[\left(H\left|\Psi^{(\alpha)}(t)\right\rangle\right)\left\langle\Psi^{(\alpha)}(t)\right| - \left|\Psi^{(\alpha)}(t)\right\rangle\left(\left\langle\Psi^{(\alpha)}(t)\right|H\right)\right] \tag{2.9}$$

$$= \frac{1}{i\hbar}\Big(H\varrho - \varrho H\Big) \tag{2.10}$$

$$= \frac{1}{i\hbar}\Big[H,\varrho\Big] \tag{2.11}$$

$$\frac{\partial\varrho}{\partial t} = \frac{1}{i\hbar}\Big[H,\varrho\Big] \tag{2.12}$$

where the second line follows from the fact that the schrödinger equation for the bra state vector $\langle\Psi^{(\alpha)}|$ is

$$-i\hbar\frac{\partial}{\partial t}\left\langle\Psi^{(\alpha)}(t)\right| = \left\langle\Psi^{(\alpha)}(t)\right|H \tag{2.13}$$

The general solution to its equation of motion is

$$\varrho(t) = e^{-\frac{iHt}{\hbar}}\varrho(0)e^{\frac{iHt}{\hbar}} = U(t)\varrho(0)U^{\dagger}(t) \tag{2.14}$$

[52]

# 3

# Materials and Methodology

This study has been carried out by using the following procedures. These are: study site and period, method of approach, materials used.

## 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 2018 to February 2019.

## 3.2 Method of Approach

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

## 3.3 Methodology

Analytical method to be employed for this study is the variational techniques. Most of the physics problems are difficult to solve analytically. Then for numerical calculations, MATLAB and MATHEMATICA SOFT WARES had been used.

# 4

# Energy eigenvalue and optical properties of ZnO cylindrical quantum wire with applied electric field

One of the most intensively explored classes of low dimensional semiconductor structures is the quantum wire. Quantum wires have attracted much attention in the theoretical and applied physics due to their potential applications in opto-electronic devices and fundamental physics investigations. One reason is that the quantum confinement of carriers in these low dimensional systems leads to the formation of discrete energy levels and the drastic changes of physical and chemical properties. Another reason is that the non linear optical properties in these system have the potential for device applications in laser amplifiers, long wavelength infrared photo detectors and high speed electro-optical modulators. In this chapter the non linear optical properties of ZnO/ZnMgO cylindrical parabolic quantum wires with applied electric field are studied.

## 4.1 Mathematical formulation of the problem

Charge carriers in a cylindrical semiconductor quantum wire with applied electric field in a two dimensional parabolic confining potential along Z-direction can be

described by the effective mass Hamiltonian.

$$H = H_r + H_z = -\frac{\hbar^2}{2m^*}\left[\frac{1}{r}\frac{\partial}{\partial r}(r\frac{\partial}{\partial r}) + \frac{1}{r^2}\frac{\partial^2}{\partial\theta^2}\right]$$

$$+\frac{1}{2}m^*\omega_0^2 r^2 - eFrcos\theta - \frac{\hbar^2}{2m^*}\frac{d^2}{dz^2}, \tag{4.1}$$

where $r = \sqrt{(x^2+y^2)}$, and $m^*$ is the effective mass of the electron in the conduction band, $\omega_0$ the the parabolic confinement frequency, e the electron charge, and F the applied electric field. $\mathbf{H}_r$ is the hamiltonian for the confined circular base (x-and y-axis)of the cylinder and $\mathbf{H}_z$ the Hamiltonian for unconfined length of the cylinder (z-axis). The electronic eigenfunctions $\psi_{nm,k}$ and eigenenergies $\varepsilon_{nm,k}$, satisfying the Schrödinger equation $H\psi_{nm,k} = \varepsilon_{nm,k}$, are given by

$$\psi_{nm,k} = \varphi_{nm}(\mathbf{r})U_c(\mathbf{r})e^{ikZ} \tag{4.2}$$

and

$$\varepsilon_{nm,k} = E_{nm} + \frac{\hbar^2 k^2}{2m^*}, \tag{4.3}$$

where k is the wave vector in the z direction and $U_c(\mathbf{r})$ is the periodic part of the Bloch function in the conduction-band bottom.$\varphi_{nm}$ and $E_{nm}$,the envelop wave functions and the transverse energies of $nm^{th}$ subband satisfying the schrödinger equation $\hat{H}\varphi_{nm} = E_{nm}\varphi_{nm}$ can be written as

$$E_{nm} = (2n + |m| + 1)\hbar\omega_0 - \frac{e^2F^2}{2m^*\omega_0^2}, \tag{4.4}$$

the ground state energy would be represented by

$$E_{00} \quad = \quad \hbar\omega_0 - \frac{e^2F^2}{2m^*\omega_0^2} \tag{4.5}$$

$$\varphi_{nm}(\rho) = \sqrt{\frac{2m^*\omega_0 n!}{\hbar(n+|m|)!}} e^{-\frac{\rho^2}{2}} \rho^{|m|}(\rho^2) \frac{1}{\sqrt{2\pi}} e^{im\psi}, \tag{4.6}$$

with

$$\rho = \sqrt{\frac{m^*\omega_0}{\hbar}\left[\left(r - \frac{eFcos\theta}{m^*\omega_0^2}\right)^2 + \left(\frac{eFsin\theta}{m^*\omega_0^2}\right)^2\right]}, \tag{4.7}$$

where n=0,1,2,..., $m = 0, \pm1, \pm2$,...,and $L^{|m|_n}$ are generalized Lagure polynomials [11].

Energy gaps for three representative confining frequencies are given as in table 4.1

| $\omega_0(\frac{1}{s})$ | $E_{00}(\times 10^{-20}J)$ | $E_{01}(\times 10^{-20}J)$ | $\Delta E = E_{10}(\times 10^{-20})$ |
|---|---|---|---|
| $4 \times 10^4$ | 4.2182 | 8.43657 | 4.2182 |
| $6 \times 10^4$ | 6.3274 | 12.6548 | 6.3274 |
| $8 \times 10^4$ | 8.4364 | 16.8729 | 8.4365 |

Table 4.1: The energy gap between first excited state and the ground state for different values of confining frequencies

The intensity of the electric field from electric field used is listed as follows with the relations

$I = 2n\epsilon_0 c|F|^2$ in table 4.2;

| No | F(v/m) | $I(w/m^2)$ |
|---|---|---|
| 1 | $1 \times 10^4$ | $1.55 \times 10^6$ |
| 2 | $2 \times 10^4$ | $6.20 \times 10^6$ |
| 3 | $3 \times 10^4$ | $1.39 \times 10^7$ |

Table 4.2: The intensity of electric field used

## 4.2 Optical properties of cylindrical quantum wire

The general expression of linear and third order nonlinear optical properties of cylindrical quantum wire determined by the compact density matrix approach and the iterative procedure. This formalism is a preferred approach due to its applicability to treat effects such as collisional broadening of atomic resonances.In order to calculate the linear and third order absorption coefficient and refractive index in cylindrical quantum wire due to optical transition, the interaction of a polarized monochromatic electromagnetic field with a quantum wire is considered.

$$\vec{E}(t) = \vec{E}e^{i\omega t} + \vec{E}^* e^{-i\omega t} \tag{4.8}$$

Due to the time dependent interaction, the time evolution of the matrix elements of the electron density operator $\hat{\varrho}$ is given by [11].

$$\frac{\partial \varrho_{nm}}{\partial t} = \frac{1}{i\hbar}\left[\hat{H} - \hat{\mu}E(t), \hat{\varrho}\right]_{nm} - \tau_{nm}(\hat{\varrho} - \hat{\varrho}^{(0)})_{nm} \tag{4.9}$$

Where $\hat{H}$ is the hamiltonian of the system in the absence of electromagnetic field $\vec{E}(t,\, \hat{\mu}) = q\hat{x}$ is the dipole moment operator along the x-axis, and $\tau_{nm}$ is the phenomenological relaxation rate, resulted by the electron-photon, electron-electron,

and other collision processes.$\varrho^{(0)}$ is the unperturbed density operator. The standard iterative method used to solve equation (4.9)is:

$$\hat{\varrho}(t) = \sum_{n=0}^{\infty} \hat{\varrho}^{(n)}, \tag{4.10}$$

with

$$\dot{\varrho}_{nm}^{(n+1)} = \frac{1}{i\hbar}[\hat{H}_0, \varrho^{(n+1)}]_{nm} - i\hbar\Gamma_{nm}^{(n+1)} - \frac{1}{i\hbar}[\mu, \varrho^{(n)}]_{nm}\vec{E}(t) \tag{4.11}$$

Moreover,the electronic polarization of the system due to this electric field $\vec{E}$ can be written as

$$P(t) = \epsilon_0\chi_{\omega}^{(1)}\vec{E}e^{i\omega t} + \epsilon_0\chi_0^{(2)}\vec{E}^2 + \epsilon_0\chi_{2\omega}^{(2)}\vec{E}^2e^{2i\omega t} + \epsilon_0\chi_{\omega}^{(3)}\vec{E}^2\vec{E}e^{i\omega t} + \epsilon_0\chi_{3\omega}^{(3)}\vec{E}^{(3)}e^{3i\omega t} + c.c \tag{4.12}$$

Where $\epsilon_0$ is the permittivity of vacuum, $\chi_{\omega}^{(1)}$,$\chi_0^{(2)}$,$\chi_{2\omega}^{(2)}$,$\chi_{\omega}^{(3)}$ and $\chi_{3\omega}^{(3)}$ are the linear, optical rectification, second harmonic generation, third order and third harmonic generation susceptibilities respectively. Concentrating on the two level system and considering the cylindrical quantum wire, centrosymmetric, we focused on linear and third order nonlinear optical properties. Using equations (4.11) and (4.12), the linear and the third order nonlinear refractive index changes are

$$\Delta n^{(1)}(\omega) = \frac{\sigma|\mu_{10}|^2}{2n_r\epsilon_0}\left[\frac{E_{10} - \hbar\omega}{(E_{10} - \hbar\omega)^2 + (\hbar\Gamma_0)^2}\right] \tag{4.13}$$

and

$$\Delta n^{(3)}(\omega) = \frac{\sigma|\mu_{10}|^2}{4n_r\epsilon_0}\frac{\nu cI}{[(E_{10} - \hbar\omega)^2 + (\hbar\Gamma_0)^2]^2}\left\{4(E_{10} - \hbar\omega)|\mu_{10}|^2 - \frac{(\mu_{11} - \mu_{00})^2}{(E_0)^2 + (\hbar\Gamma_0)^2}\right.$$

$$\left.\times\left((\hbar\Gamma_0)^2(2E_{10} - \hbar\omega) - (E_{10} - \hbar\omega)\left[(E_{10} - \hbar\omega)E_{10} - (\hbar\Gamma_0)^2\right]\right)\right\} \tag{4.14}$$

Where $\sigma$ is the carrier density, $\mu_{nm} = |\langle\psi_n|q\hat{x}|\psi_m\rangle|$,(n,m=1,2) are the matrix elements of the dipole moment,$\psi_n$ and $\psi_m$ are the eigen functions, $E_{nm} = E_n - E_m$is the energy differences between these states, $n_r$ is the refractive index, $\nu$ is the permeability,c is the speed of light and I is the intensity of the electric field. The first and third order optical absorption coefficients can be described as

$$\beta^{(1)}(\omega) = \omega\sqrt{\frac{\nu}{\epsilon}}\frac{|\mu_{10}|^2\sigma\hbar\Gamma_0}{(E_{10}-\hbar\omega)^2+(\hbar\Gamma_0)^2} \tag{4.15}$$

$$\beta^{(3)}(\omega) = -\omega\sqrt{\frac{\nu}{\epsilon}}\left(\frac{I}{2\epsilon_0 n_r c}\right)\frac{|\mu_{10}|^2\sigma\hbar\Gamma_0}{[(E_{10}-\hbar\omega)^2+(\hbar\Gamma_0)^2]^2}$$

$$\times\left\{4|\mu_{21}|^2 - \frac{|\mu_{22}-\mu_{11}|^2[3E_{10}^2 - 4E_{10}\hbar\omega + \hbar^2(\omega^2-\Gamma_0^2)]}{E_{10}^2+(\hbar\Gamma_0)^2}\right\} \tag{4.16}$$

[11]

## 4.3 Numerical results and discussions

The calculation is made for linear and third order nonlinear index of refraction and absorption coefficient of ZnO quantum wire with carrier density $\sigma = 3\times10^{22}m^{-3}$, effective mass $m^* = 0.21m_e$, the background dielectric constant $\epsilon$ = 8.5, the refractive index $n_r$ = 2.92, the permeability $\nu = 4\pi\times10^{-7}N/A^2$, the speed of light in vacuum $c = 3\times10^8$m/s, the relaxation frequency $\tau_0 = 2\times10^{11}\frac{1}{s}$, the applied electric field $F = 1\times10^4$v/m, $2\times10^4$v/m, $3\times10^4$v/m,The confining energy $E_{10} = 4.216\times10^{-20}$J for the linear one and $E_{10} = 4.216\times10^{-20}$J, $4.427\times10^{-20}$J,and $4.848\times10^{-20}$J energies for the nonlinear one. The radius of the cylindrical quantum wire we are interested in is 1.2nm from confining frequency $\omega_0$ of 0.4THz

In the next session four graphs that explain the mentioned property of the linear and third order nonlinear index of refraction and absorption coefficient would be presented. .

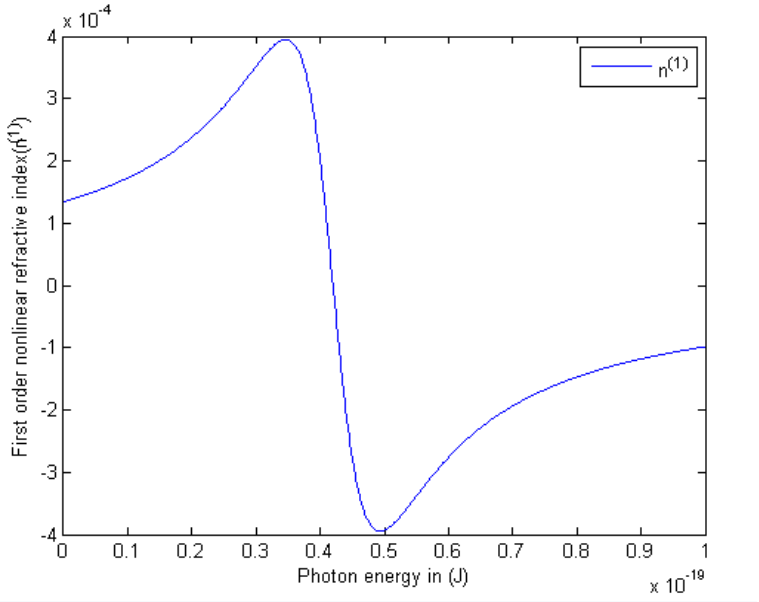


Figure 4.1: The linear refractive index $\Delta n^{(1)}(\omega)$versus photon energy $(\hbar\omega)$ in (J)for ZnO Cylindrical quantum wire

Figure.4.1 shows the linear refractive index $(n^{(1)}(\omega))$ which is plotted as a function of photon energy $\hbar\omega$ with the dipole moment $\mu_{10}$=1.1$\times 10^{-28}$c-m and relaxation time $\tau_0$= 2$\times 10^{11}\frac{1}{s}$ using Eq 4.15. From the relation, (Eq 4,15), and also the figure 4.1 one can conclude the linear index of refraction is not electric field intensity dependent. It is larger in magnitude.

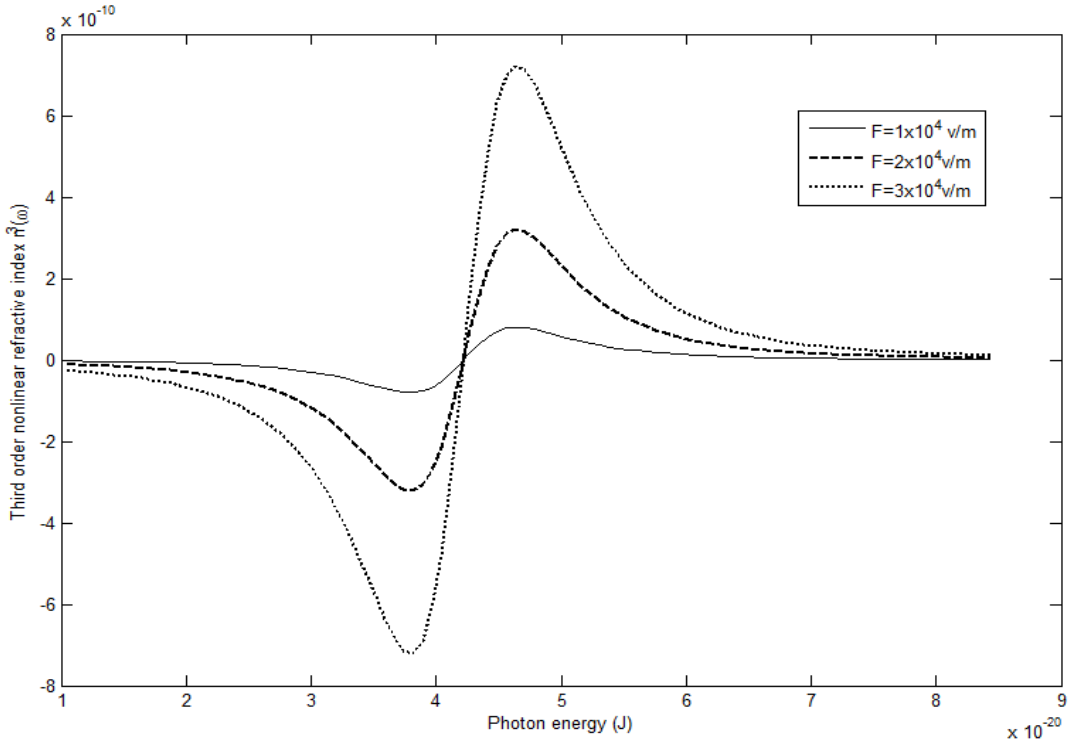


Figure 4.2: The third order nonlinear refractive index versus photon energy for different values of magnitude of applied electric field

Figure 4.2 shows the third order nonlinear refractive index plotted as a function of photon energy $(\hbar\omega)$, with intensity (I=1.550$\times 10^6$, 6.202$\times 10^6$and 1.395$\times 10^7 W/m^2$), the relaxation time $\tau_0$=2$\times 10^{11}\frac{1}{s}$ ,and the with respective dipole moment $\mu_{10}$=1.1 $\times$ $10^{-28}$, 1.2$\times 10^{-28}$, and 1.3$\times 10^{-28}$c-m. using Eq 4.16.

It is very small in comparison to the linear one. From this figure (Fig 4.2) one can deduce that as the magnitude of the applied electric field intensity increases the magnitude of third order nonlinear refractive index increases.

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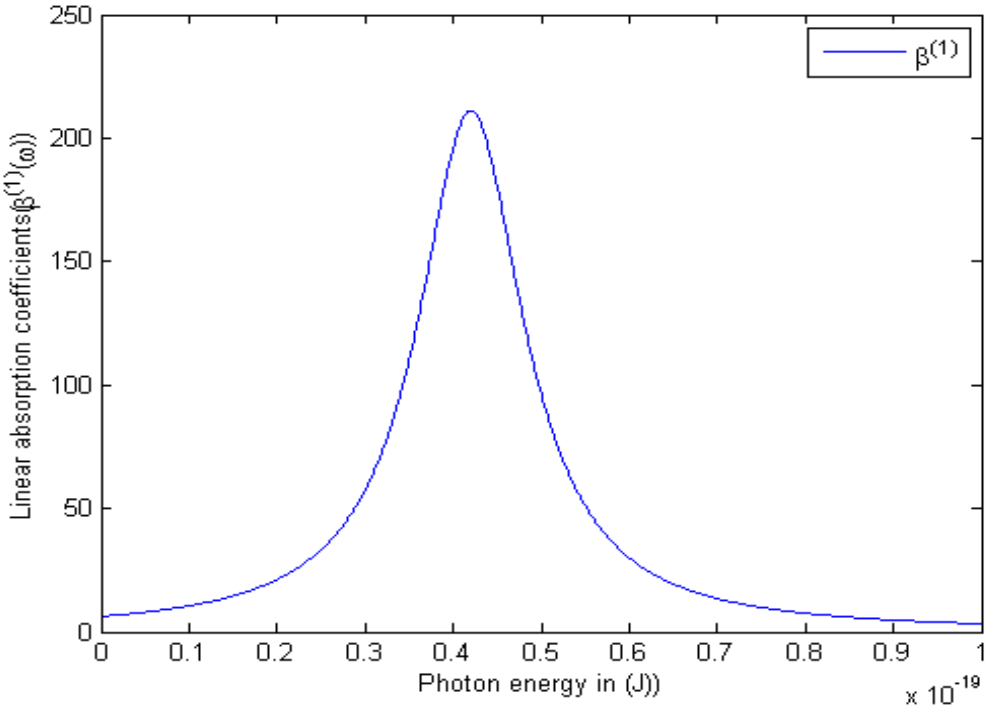


Figure 4.3: The linear absorption coefficient as a function of photon energy $(\hbar\omega)$ in (J)for ZnO Cylindrical quantum wire

Figure 4.3 shows the graph of lineaer absorption coefficient of ZnO quantum wire versus photon energy. Since the linear absorption coefficient is not related to intensity it is not intensity dependent and it is large in magnitude.

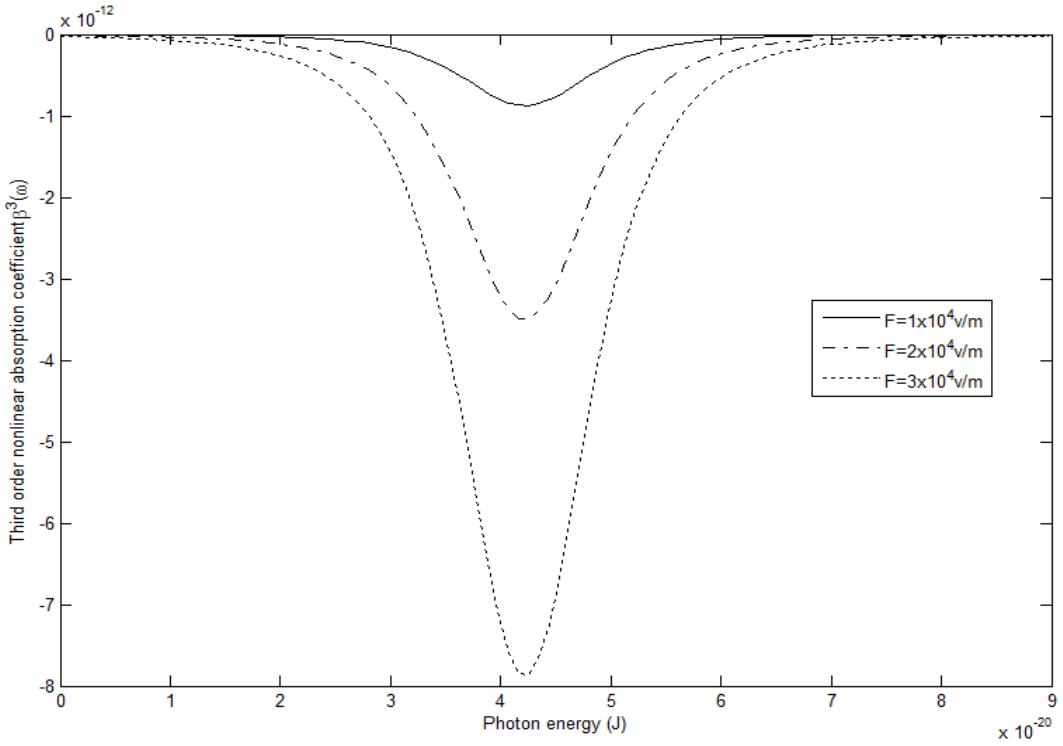


Figure 4.4: The third order nonlinear absorption coefficient versus photon energy values of magnitude of electric field

Figure 4.4 shows the graph of third order nonlinear absorption coefficient of ZnO versus photon energy. It is very small in magnitude. As the magnitude of the applied electric field intensity increases the third order nonlinear absorption coefficient increases.

# 5

# Conclusion

In this study, first the ground and excited state energies of ZnO cylindrical quantum wire in parabolic confining potential with the applied electric field are calculated. Then using the obtained energy eigen values, the first order and third order refractive indexes and absorption coefficients are determined with the help of density matrix formalism. The result shows that the magnitude of the linear refractive index is large compared to the magnitude of third order nonlinear refractive index. The linear refractive index does not depend on the intensity. However, the magnitude of the third order refractive index depends on the optical intensity and increases as the optical intensity and increases. Moreover, the magnitude of the third order absorption coefficient is too small as compared to the linear absorption coefficient. In addition to this, the third order absorption coefficient depends on the optical intensity and its magnitude is amplified as the optical intensity increases. This intensity dependent nonlinear optical property has a potential application in nonlinear optical devices such as lasers, photodetectors and electro-optical modulators.

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