OPTICAL RECTIFICATION AND SECOND ORDER NONLINEAR OPTICAL PROPERTIES OF ZINC OXIDE PARABOLIC QUANTUM WELL WITH APPLIED ELECTRIC FIELD

Eliyas Mohammed Jifar

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Supervisor: _____
Dr. Menberu Mengesha

Readers: _____
Solomon H/Mariam

_____

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Author: **Eliyas Mohammed Jifar**

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To my family.

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Abstract

In this thesis, the Hamiltonian and wave functions of parabolic quantum wells with applied electric field are developed. Then the *Schrödinger* equation is solved analytically and numerically for determining the energy eigenvalue using variational method. The energy eigenvalues are decreasing with the increment of an applied electric field. However, the energy spacing between two states are constant. By using the compact-density matrix formalism and iterative procedure, the optical rectification $\chi_0^{(2)}$ is calculated for the parabolic quantum wells. Numerical results show that, optical rectification (OR) coefficient is strongly affected by the magnitude of applied electric field. The magnitude of optical rectification was decreasing with the increment of the magnitude of applied electric field F. Furthermore, the phenomenological damping constant has a great influence on second-order nonlinear optical rectification (OR). With increase of damping constant, the magnitude of optical rectification decreases. Again, the maxima of optical rectification shifts towards the higher energy, as a confinement frequency of parabolic quantum well increases.

Chapter 1

1. Introduction

1.1 Back ground of the study

A quantum well is a particular kind of heterostructure in which one thin "well" layer is surrounded by two "barriers" layers. Quantum wells are one example of heterostructures - structures made by sandwiching different materials, usually in layers, and with the materials joined directly at the atomic level [1]. The quantization of the particle motion occurs in one direction, while the particle is free to move in other two directions. The optical properties of quantum wells specially optical absorption are strongly affected by the material and as well as the external factors such as incidence optical intensity, electric and magnetic field, laser fields, temperature and pressure [2]. The properties of quantum wells make them unique materials in the field of non-linear optics and electro-optics.

Parabolic quantum wells are very interesting structure both from fundamental and technological points of view [3]. Parabolic quantum well is a symmetrical quantum system, while semi-parabolic quantum well is asymmetric quantum system. Parabolic quantum wells have the particular property of equally spaced electronic states, which makes possible an accurate determination of the band offsets parameters. Recently,

parabolic quantum wells have been implemented to study nonlinear optical properties. ZnMgO-ZnO-ZnMgO is quantum well made from zinc oxide (a narrow band gap material) sandwiched between two barriers of zinc magnesium oxide (a wide band gap material). Bearing this idea, we will study the influence of electric field on the optical rectification and second-order nonlinear optical properties of ZnMgO-ZnO-ZnMgO parabolic quantum wells.

1.2 Statement of the problems

Quantum well has found widespread use in light emitting diodes and laser diode applications for a number of years now. In quantum wells, quantization of the particle motion occurs in one direction, while the particle is free to move in other two directions. The application of quantum well structures to semiconductor laser diodes has received considerable attention because of physical interest as well as its superior characteristics such as low threshold current density, low temperature dependence of threshold current, lasing wavelength tenability and excellent dynamic properties [4]. By controlling the width of the quantum wells, one can modify the electron and hole wave functions, which leads to the modification of materials parameters. Heterostructures are the building blocks of many of the most advanced semiconductor devices presently being developed and produced. Heterostructures are able to improve the performance of semiconductor because they permit the device designer to locally modify the energy band structure of the semi-conductor and so control the motion of the charge carriers [5]. Optical measurements provide a direct evidence for the low dimensional behavior of electrons and holes in the quantum well. The Fermi-edge singularity was predicted theoretically and experimentally demonstrated for an

expressive number semiconductor heterostructure [6]. The Fermi-edge singularity leads to a strong enhancement of the oscillator strength for the transitions involving states at the Fermi energy, giving rise to a sharp excite on like emission in the optical spectral. In this thesis work, we seek to calculate the energy eigenvalue of parabolic quantum well and to understand the optical rectification and second order nonlinear optical properties of ZnMgO-ZnO-ZnMgO parabolic quantum well with applied electric field.

Research Questions

1. How can we calculate the energy eigenvalue of parabolic quantum well varying the magnitude of applied electric field?

2. How can we study the optical rectification of ZnMgO-ZnO-ZnMgO parabolic quantum wells in applied electric field?

3. How can we describe the second order nonlinear optical properties of ZnMgO-ZnO-ZnMgO parabolic quantum wells in applied electric field?

1.3 Objective

1.3.1 General objective

- To study the optical rectification and second order nonlinear optical properties of ZnMgO-ZnO-ZnMgO parabolic quantum wells with applied electric field.

1.3.2 Specific objective

- To calculate the energy eigenvalue of parabolic quantum well varying the magnitude of applied electric field.

- To study the optical rectification of ZnMgO-ZnO-ZnMgO parabolic quantum wells in applied electric field
- To describe the second order nonlinear optical properties of ZnMgO-ZnO-ZnMgO parabolic quantum wells in applied electric field.

1.4 Significance of the study

This study helps to calculate, the energy eigenvalue of parabolic quantum well with applied electric field. It also helps to understand, the lowest-order nonlinearity, such as second-order nonlinear optical property.

1.5 Limitation of the study

Due to time constraint, the scope of the study is limited to the calculation of energy eigenvalue of parabolic quantum well with applied electric field and study, the properties of second-order nonlinear optical properties; such as optical rectification and second-harmonic generation.

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Chapter 2

2. Literature Review

2.1 Nonlinear Optics

Nonlinear optics is the study of phenomena that occurs as a sequence of modification of the optical properties of a material system by presence of light [7]. Nonlinear optics is also the area of optics that studies the interaction of light with matter in the regime where the response of the material to the applied electromagnetic field is nonlinear in the amplitude of this field. In nonlinear optics, optical properties are dependent of light (light interact with nonlinear medium). Nonlinear optics takes place, when a material interacts with an intense light. Nonlinear optics is the branch of optics that describes the behavior of light in nonlinear media. Nonlinear media is media in which the dielectric polarization **P** responds nonlinearly to the electric field **E** of the light.

When you shine light at one frequency on nonlinear optical medium, you get out light with twice frequency and when you shine light at one frequency on linear optical medium, you get out light with the same frequency.

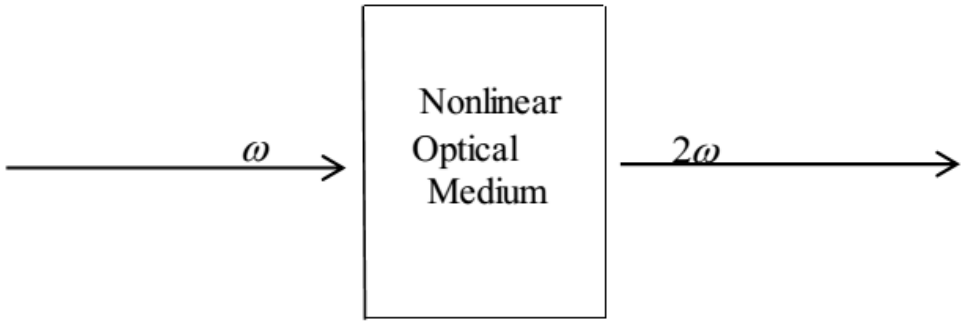

Figure 2.1: *Nonlinear medium produce double frequency.*

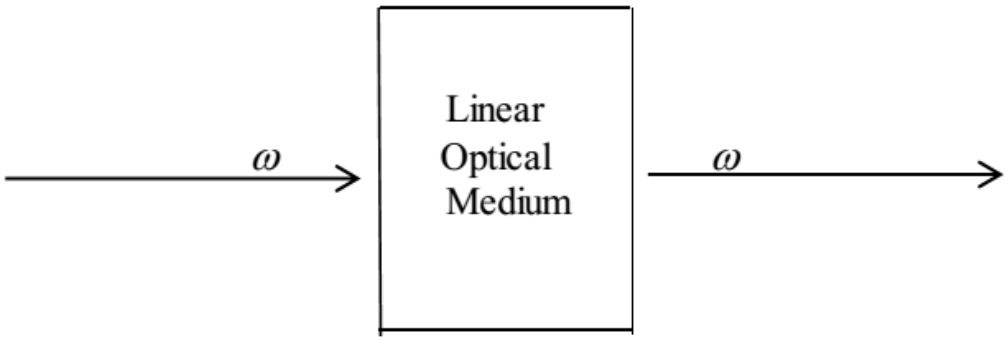

Figure 2.2: *Linear medium produce the same frequency.*

Both linear and nonlinear optical effects can be understood as resulting from the interaction of the electric field component of electromagnetic radiation with the charged particles of the material. An applied electric field moves positively charged particles in the direction of the field and negative charges in the opposite direction. Nonlinear optical phenomena are "nonlinear" in the sense that they occurs when the response of material system to an applied optical field depends in a nonlinear manner on the strength of the optical field. 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 $\mathbf{E}(t)$, of a material system depends on the strength $\mathbf{P}(t)$ 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 manner that often be described by the relation ship

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

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) by expressing the polarization $\mathbf{P}(t)$ as a power series in the field strength $\mathbf{E}(t)$ as

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

The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second and third order nonlinear optical susceptibility respectively. Second-order susceptibility is a nonlinear property of the material describing how the polarization develops due to the product of two field components. We shall refer to $\mathbf{P}^{(2)}(t) = \epsilon_0 \chi^{(2)} \mathbf{E}^2(t)$ as the second-order nonlinear polarization and $P^{(3)}(t) = \epsilon_0 \chi^{(3)} \mathbf{E}^3(t)$ as the third-order nonlinear polarization [7]. Second-order nonlinear optical interactions can occur only in non centrosymmetric crystals. That is, in crystal that does not display inversion symmetry. Since, liquid, gases, amorphous solid (such glass), and even many crystals display inversion symmetry, $\chi^{(2)}$ vanishes identically for such media, and consequently such materials cannot produce second-order nonlinear optical interactions. On the other hand, third-order nonlinear optical interactions (i.e. those described by a $\chi^{(3)}$ susceptibility) can occurs for both centrosymmetric and non centrosymmetric media. The most usual procedure

for describing nonlinear optical phenomena is based on expressing polarization $\mathbf{P}(t)$ in terms of the applied electric field strength $\mathbf{E}(t)$ as we have done in Eq.(2.1.2). The reason why the polarization plays a key role in the description of nonlinear phenomena is that time-varying polarization can act as the source of new components of the electromagnetic field. The wave equation in nonlinear optical media often has the form

$$\nabla^2\mathbf{E} - \frac{n^2\partial^2\mathbf{E}}{c^2\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2\mathbf{P}^{NL}}{\partial t^2} \tag{2.1.3}$$

Where n is the usual linear refractive index and c is speed of light in vacuum we can interpret this expansion as an inhomogeneous wave equation in which the polarization $\mathbf{P}^{NL}$ associated with the nonlinear response drives the electric field $\mathbf{E}$. Since $\frac{\partial^2\mathbf{P}^{NL}}{\partial t^2}$ is the measure of the acceleration of the charges that constitute the medium, this equation is consistent with Larmor's theorem of electromagnetism which states that accelerated charges generate electromagnetic radiation.

The general principle here is that carriers created optically will change the field across the quantum wells, hence changing their optical properties through the Quantum Confined Stark Effect [8]. Nonlinearities are certainly non-local (because a light beam at one place can influence the optical properties at another), and for this reason cannot strictly be described by the usual local nonlinear susceptibility formalism. They are similar to those involved in photorefractive materials where displacement of charges toward traps generates large electric field which in turn modifies the optical properties through the normal electro-optic effect. Consequently, in order to obtain the enhanced second-order nonlinear optical susceptibility in quantum wells, externally applied electric fields are used to remove the symmetry or the quantum wells structures are produced with a built in asymmetry using advanced material

growing technology. Among the nonlinear optical properties, it is attracted much attention to the second-order nonlinear optical properties, such as optical rectification and second-harmonic generation. It is because the second-order nonlinear processes are the simplest and the lowest-order nonlinear effects, and the magnitude of these second-order nonlinear coefficients are usually stronger than that of the higher-order ones, as the symmetry of quantum system is broken [9, 10]. Recently, nonlinear optical properties in semiconductor quantum wells are of considerable interest because of their relevance for studying practical applications. Much special has been paid to the second-order nonlinear optical properties, such as electro-optic effect, optical rectification, second-harmonic generation and so on, because the second-order nonlinear procedures are the simplest and the lowest-order nonlinear procedures, and the magnitudes of the second-order nonlinear procedures are usually stronger than those of the higher-order nonlinear procedures when the quantum systems have significant asymmetry [11]. The nonlinear response of the medium produces higher harmonics in the polarization.

2.1.1 Electro-Optic Effect

The electro-optic is the change in refractive index of material induced by the presence of a static (or low-frequency) electric field [7]. In some materials, the change in refractive index depends linearly on the strength of the applied electric field. This change is known as the linear electro-optic effect or Pockels effect. On the other hand, the refractive index changes in proportional to the applied electric field, in which case the effect is known as the linear electro-optic effect or the Pockels effect. The Pockels effect has the linear relationship to the applied field. This is principally a second-order nonlinear optical phenomenon. Second-order nonlinear optical processes are

electric-dipole forbidden in centrosymmetric media [12]. Second-order nonlinearities are nonzero only in noncentrosymmetric systems. That means, a semiconductor quantum well having symmetric potential profile will provide very small second-order nonlinearities. Second-order nonlinear optical properties describe the coupling interaction between two electric fields and crystals [13]. In simple terms, the Pockels effect describes the linear dependence of the refractive index on an applied field. The linear electro-optic effect can be descried in terms of a nonlinear polarization given by

$$\mathbf{P}_i(\omega) = 2\epsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega = \omega + 0)\mathbf{E}_j(\omega)\mathbf{E}_k(0) \tag{2.1.4}$$

Since the linear electro-optic effect can be described by a second-order nonlinear susceptibility. It follows that a linear electro-optic effect can occurs only for materials that are non centrosymmetric. In centrosymmetric materials (such as liquids and glasses), the lowest-order change in the refractive index depends quadratically on the strength of the applied static (or low-frequency) field. This effect is known as the Kerr electro-optic effect. On the other hand, the refractive index changes in proportional to the square of the applied electric field, in which case the effect is known as the quadratic electro-optic effect or the Kerr effect. The Kerr effect has a parabolic relationship to the applied field. Both the linear quadratic electro-optic effect can be used effectively in various optical devices. In Figure 2.3, the refractive index as a function of applied field n(E) shows the linear and quadratic electro-optic effect.

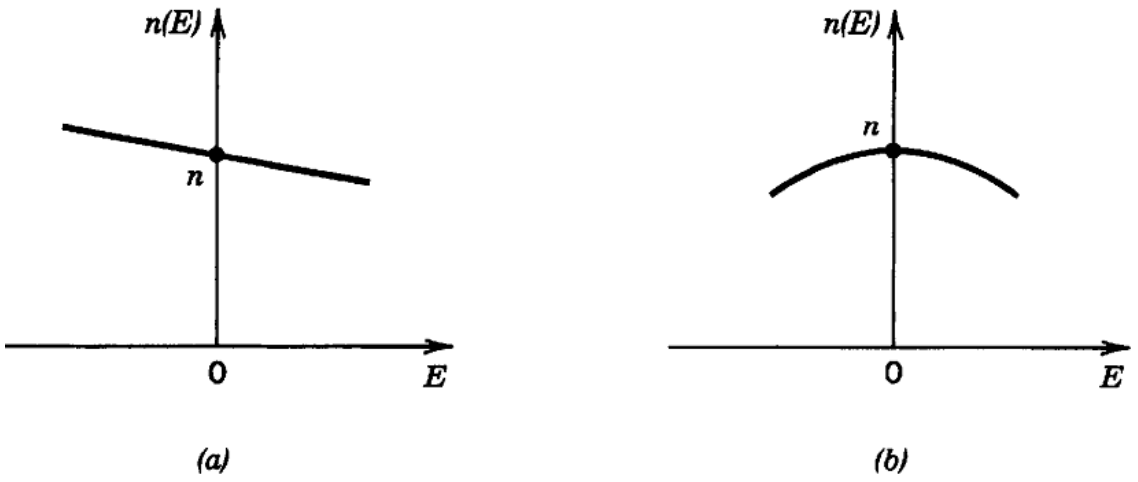

Figure 2.3: *Dependence of the refractive index on the electric field: (a) Pockels medium; (b) Kerr medium.*

Kerr electro-optic effect can be described in terms of a nonlinear polarization given by

$$\mathbf{P}_i(\omega) = 3\epsilon_0 \sum_{jkl} \chi^{(3)}_{ijkl}(\omega = \omega + 0 + 0)\mathbf{E}_j(\omega)\mathbf{E}_k(0)\mathbf{E}_l(0) \tag{2.1.5}$$

Both the linear and quadratic electro-optic effects can be used effectively in various optical devices. Nonlinear and electro-optic properties of materials are linked to the modification of their optical constants by high-intensity optical fields or by DC electric fields respectively [14]. Generally, the electro-optic effect is the term used to describe the changes in the optical properties of the material due to the presence of an electric field.

2.1.2 Optical Rectification

Among the nonlinear optical properties, it attracted much attention to the second-order nonlinear optical properties, such as optical rectification and second-harmonic

generation [8]. Therefore, optical rectification is a nonlinear optical process that consists of the generation of the quasi-DC polarization in a nonlinear medium at the passage of an intense optical beam. The DC polarization results from the rectification of the incident optical electric field by the second-order nonlinear electric susceptibility of the material. Optical rectification is the creation of a static electric field in the materials due to the incident optical field. Generally, optical rectification refers to the development of a DC or low-frequency polarization when intense laser beams propagate through a crystal. Or optical rectification is nonlinear optical process that generates a quasi-DC polarization in nonlinear medium by passing an intense beam of light. So, optical rectification occurs, when an intense light beam transmits through a crystal. Optical rectification is observable only in noncentrosymmetric systems. That means, optical rectification occurs only in crystals that are not centrosymmetric. However, optical rectification of laser light by centrosymmetric crystals is possible if the symmetry is broken by a strong electric field. Optical rectification and the linear electro-optic effect are second-order nonlinear optical effects $\mathbf{P}_2^{nl}$ and described by the $\mathbf{P}_2^{nl} = \chi_2 \mathbf{E}^2$ terms in the expansion. In this case, the second-order nonlinear polarization $\mathbf{P}_2^{nl}$ consists of a dc polarization $\chi_2 \mathbf{E}^2$.

2.1.3 Second-Harmonic Generation

One important application of second-harmonic generation is its use as exacting diagnostic of the surface properties of optical materials. The surface of a material clearly lacks inversion symmetry, and thus, second-harmonic generation can occurs at the surface of a material of any symmetry group [7]. Second-harmonic generation is just a special sum frequency generation where an optical wave interacts with itself to generate the sum frequency. Second-harmonic generation (also called frequency

doubling) is a nonlinear optical process, in which photons with the same frequency interacting with a nonlinear material are effectively "combined" to generate new photons with twice the energy, and therefore twice the frequency and half the wavelength of the initial photons. In second-harmonic generation, a pump wave with a frequency of ω generates a signal at the frequency 2ω as it propagates through a medium with a quadratic nonlinearity proportional to the macroscopic second-order susceptibility $\chi^{(2)}$. Therefore, the process of second-harmonic generation involves the interaction of two waves at frequency ω to produce a wave with the frequency 2ω. Among the nonlinear phenomena existing in nature, the main role is played by second-harmonic generation. Second-harmonic generation, as an even-order nonlinear optical effect, is only allowed in media without inversion symmetry. That means in centrosymmetric materials, second-harmonic generation cannot be demonstrated, because of the inversion symmetry. It is also important to note that since all even-order nonlinear susceptibilities $\chi^{(n)}$ vanish in centrosymmetric media. Because of its properties, second-harmonic generation finds many different and interesting applications. As the applied electric field becomes strong, the second harmonic generation susceptibility in semi-parabolic and parabolic quantum well systems nearly increases linearly [11]. Furthermore, when the effective widths of the semi-parabolic and parabolic quantum wells are the same, the second-harmonic generation susceptibility in semi-parabolic quantum well is larger than that in parabolic quantum well, which means that the semi-parabolic quantum well is a model of very promising candidates for the second-order nonlinear optical properties. Second-harmonic generation occurs in three types, denoted 0, I and II. In type 0 second-harmonic generation, two photons having extraordinary polarization with respect to the crystal will combine to form a single

photon with double the frequency/ energy and extraordinary polarization. In type I second-harmonic generation, two photons having ordinary polarization with respect to the crystal will combine to form one photon with double the frequency and extraordinary polarization. In type II second-harmonic generation, two photons having orthogonal polarizations will combine to form one photon with double the frequency and extraordinary polarization. For a given crystal orientation, only one of these types second-harmonic generation occurs.

2.2 Quantum Wells

A quantum well is a particular kind of heterostructure in which one thin "well" layer is surrounded by two "barriers" layers [1]. Quantum wells are one example of heterostructures - structures made by joint different materials, usually in layers, and with the materials joined directly at the atomic level. If one makes heterostructure with sufficiently thin layers, quantum interference effects begin to appear prominently in the motion of the electrons [5]. The simple structure in which these may be observed is a quantum well, which simply consist of a thin layer of a narrower-gap semiconductor between thicker layers of a wider-gap material. Quantum well is two-dimensional structure and quantization of a particle motion occurs in one direction, while the particle is free to move in the other two directions. In a quantum well, the electrons and holes are still free to move in the directions parallel to the layers; hence, we do not really have discrete energy states for electrons and holes in quantum wells.

The most general and surprising features of the optical properties of quantum well is the strength of the intrinsic optical effects as compared to bulk optical properties: in many circumstances one measures features of comparable size for single quantum

No	Systems	D_f	D_c
1	Bulk	3	0
2	Quantum well	2	1
3	Quantum wire	1	2
4	Quantum dot	0	3

Table 2.1: *The number of degrees of freedom D_f and the confinement D_c of the four basic dimensionality systems.*

well of $\approx 100A^0$ as for bulk samples of thickness of the order of absorption length, a few $100A^0$ [14]. For the bulk susceptibility, the nonlinear effect is not very large because of the symmetry of the crystal structure. The properties of quantum wells make them unique materials in the field of nonlinear optics and electro-optics.

Quantum wells, leads to better performance in optical devices such as laser diodes. As a result, quantum wells are in wide use in diode lasers, including red lasers for DVDs and laser pointers, infrared lasers in fiber optic transmitters or in blue lasers. They are also used to make high electron mobility transistor, which are used in low-noise electronics.

The dimensionality refers to the number of degree of freedom in the electron momentum; in fact, with in a quantum wire, the electron is confined across two directions, rather than just the one in a quantum well, and so therefore, reducing the degree of freedom to one. In a quantum dot,the electron is confined in all three dimensions, thus reducing the degrees of freedom to zero.

Four basic dimensionality systems are shown in below table.

2.3 Zinc Oxide (ZnO)

Zinc oxide is an inorganic compound with the formula ZnO. ZnO is a white powder that is insoluble in water. Zinc oxide is a II-VI compound semiconductor whose

ionicity resides at borderline between the covalent and ionic semiconductor [13]. Compendiums dealing with optical properties of ZnO and to some extent its alloys from far infrared to vacuum ultraviolet including phonons, plasmons, dielectric constant and refractive indices are available in the literature. Zinc oxide is an important II-VI semiconductor material with a wide direct band gap, which has a large exciton binding energy at room temperature, even larger than the ionization energy at the same temperature [15]. Internal electric fields were found to appear naturally in strained group III-nitride-based quantum structures grown. Despite the fact that theoretical calculations of the polarization properties in ZnO and (Zn, Mg)O predict larger spontaneous and piezoelectric constants than for GaN-based systems, the first experimental results reported on ZnO/(Zn, Mg)O quantum wells do not mention the presence of such a field. Zinc oxide is a II-VI semiconductor compound that has numerous potential application [16]. Zinc oxide crystallizes preferentially in the hexagonal structure and its wide band gap of 3.27eV is close to that of gallium nitride (GaN-3.42eV). The band gap energy at room temperature increases from 3.27eV for ZnO to 3.87eV for 33% magnesium in the alloy, with a fairly small lattice mismatch between the binary and ternary compounds. Zinc oxide and related compounds have gained a renewed interest for optoelectronic applications owing in part to the fact that ZnO/(Zn, Mg)O quantum wells have been successfully grown by laser-assisted molecular-beam epitaxy. Among II-VI wide band gap semiconductors, zinc oxide is interesting materials for its potential applications in optoelectronic devices [17]. Moreover, a large built-in internal field due to spontaneous and piezoelectric polarizations exists in Wurtzite Mg based on ZnO quantum wells. Hence, it is a promising semiconductor with the direct band gap around 3.3eV with some applications in short

wavelength optical devices and long life time operating devices. To understand the fundamental optical properties in optoelectronic devices, it is required to know the excitonic properties. ZnO has a larger exciton binding energy, around 60meV. ZnO is an interesting materials with applications spanning from a simple and well-known to the highly advanced and sophisticated [18]. Its wide band gap and high absorptivity have made it useful as UV-absorbing additive in everything from sunscreens, advanced plastics and rubber. It is also used in pigments and lubricants.

Multiple quantum well structures consist of more than one well so that more carriers can be injected into the system [19]. Recently, much effort has been devoted towards the investigation and fabrication of symmetric ZnO/ZnMgO multiple quantum wells for ultraviolet light-emitting applications. Nowadays ZnO is widely used for production of blue-ultraviolet light-emitted device and detectors [15]. ZnO quantum well structures have been fabricated to realize high efficiency luminescence in light-emitted devices. Compared to square quantum wells, the optical transition between the size-quantized electron sub bands are easier to implement and the optical nonlinearities are stronger in the parabolic quantum wells.

2.4 Parabolic Quantum Wells

Parabolic quantum wells are very interesting structure both from fundamental and technological points of view [3]. Parabolic quantum is a symmetrical quantum system, while semi-parabolic quantum well is asymmetric quantum system. In these structures it is possible to form an almost homogeneous electron gas within a three-dimensional space that moves in a uniformly distributed background of positive charge. Moreover, parabolic quantum wells have the particular property of equally

spaced electronic states, which makes possible an accurate determination of the band offsets parameters. From the technological points of view, parabolic quantum wells can be used as polarization insensitive electron-absorptive modulators and far-infrared resonant tunneling devices. Finally, parabolic quantum wells demonstrated to be a very good candidate for the electric control of the electron spin, essential for the development of spintronic devices.

In particular, the parabolic quantum well, aside to the square one, is one of the most studied systems both from fundamental and technological points of view [20]. This is, partially, because of its unique properties such as: equally spaced, electronic spectrum, radiate transitions at the same oscillator frequency, interaction with light at the oscillator frequency irrespective of electron-electron interactions, i.e., independence on the number of electrons in the well and on an electric field applied across the well, and so called generalized Kohn theorem. The generalized Kohn theorem states that, electrons residing in a spatially parabolic potential well and in parabolic bands in k space absorb radiation at the bare harmonic oscillator frequency of the empty parabola irrespective of electron-electron interaction or the number of electrons in the well [21]. With the exception of the case of a superlattice superimposed on the parabolic quantum well (where a mass-shifted generalized Kohn theorem response is observed), the wells with deviations from perfect parabolicity display a violation of the theorem qualitatively and quantitatively consistent with a uniform radiation field now coupling not only to the center of mass oscillation of the electron gas, but also to its internal oscillations. Recently, parabolic quantum wells have been implemented to study nonlinear optical properties, quantum hall effect, charge and spin oscillators in three-dimensional gases and magnetic properties for the spin electronics.

2.5 Density matrix formalism of the second-order susceptibility

In this section we calculate the second-order (i.e., $\chi^{(2)}$) susceptibility of an atomic system [7]. The density operator is given by:

$$\hat{\rho} = |\psi\rangle\langle\psi|$$

$$\dot{\rho} = \frac{d\hat{\rho}}{dt} = \frac{d}{dt}|\psi\rangle\langle\psi| + |\psi\rangle\frac{d}{dt}\langle\psi|$$

If a quantum-mechanical system (such as an atom) is known to be in a particular quantum-mechanical state, we can describe all of the physical properties of the system in terms of the wave function ψ appropriate to this state. This wave function obeys the Schrödinger equation

$$i\hbar\frac{d}{dt}|\psi\rangle = \hat{H}|\psi\rangle$$

but

$$\frac{d}{dt}|\psi\rangle = \frac{-i}{\hbar}\hat{H}|\psi\rangle$$

and

$$\frac{d}{dt}\langle\psi| = \frac{i}{\hbar}\hat{H}\langle\psi|$$

$$\begin{aligned}
\dot{\rho} = & \frac{-i}{\hbar}\hat{H}|\psi\rangle\langle\psi| + |\psi\rangle\frac{i}{\hbar}\hat{H}\langle\psi| \\
= & \frac{-i}{\hbar}\hat{H}|\psi\rangle\langle\psi| + \frac{i}{\hbar}|\psi\rangle\langle\psi|\hat{H} \\
= & \frac{-i}{\hbar}\hat{H}\hat{\rho} + \frac{i}{\hbar}\hat{\rho}\hat{H} \\
= & \frac{-i}{\hbar}[\hat{H}, \hat{\rho}]
\end{aligned}$$

Therefore, the density matrix equation of motion with the phenomenological inclusion of damping is given by:

$$\dot{\rho}_{nm} = \frac{-i}{\hbar}[\hat{H}, \hat{\rho}]_{nm} - \gamma_{nm}\left(\rho_{nm} - \rho_{nm}^{(eq)}\right)$$

From the perturbation expansion $\rho_{nm}^{(1)}(t) = \int_{-\infty}^{t} \frac{-i}{\hbar}\left[\hat{V}(t'), \hat{\rho}^{(0)}\right]_{nm} e^{(i\omega_{nm}+\gamma_{nm})(t'-t)}dt'$, the general result for the second-order correction to $\hat{\rho}$ is given by:

$$\begin{aligned}
\rho_{nm}^{(2)} &= \int_{-\infty}^{t} \frac{-i}{\hbar}\left[\hat{V}, \hat{\rho}^{(1)}\right]_{nm} e^{(i\omega_{nm}+\gamma_{nm})(t'-t)}dt' \\
&= \int_{-\infty}^{t} \frac{-i}{\hbar}\left[\hat{V}, \hat{\rho}^{(1)}\right]_{nm} e^{(i\omega_{nm}+\gamma_{nm})t} e^{(i\omega_{nm}+\gamma_{nm})t'}dt' \\
&= e^{-(i\omega_{nm}+\gamma_{nm})t} \int_{-\infty}^{t} \frac{-i}{\hbar}\left[\hat{V}, \hat{\rho}^{(1)}\right]_{nm} e^{(i\omega_{nm}+\gamma_{nm})t'}dt'
\end{aligned} \tag{2.5.1}$$

The interaction Hamiltonian $\hat{V}(t)$ is given by

$$\hat{V}(t) = -\hat{\mu}.\mathbf{E}(t) \tag{2.5.2}$$

we represent the applied fields as:

$$\mathbf{E}(t) = \sum_{p} \mathbf{E}(\omega_p)e^{-\omega_p t} \tag{2.5.3}$$

The commutator in Eq. (2.5.1) can be expressed as

$$\left[\hat{V}, \hat{\rho^{(1)}}\right]_{nm} = \sum_{v}\left[V_{nv}\rho_{vm}^{(1)} - \rho_{nv}^{(1)}V_{vm}\right] \tag{2.5.4}$$

Inserting Eq. (2.5.2) into Eq. (2.5.4), we have,

$$\begin{aligned}
\left[\hat{V}, \hat{\rho^{(1)}}\right]_{nm} &= \sum_{v}\left[-\mu_{nv}\mathbf{E}(t)\rho_{vm}^{(1)} + \rho_{nv}^{(1)}\mu_{vm}.\mathbf{E}(t)\right] \\
&= -\sum_{v}\left[\mu_{nv}\rho_{vm}^{(1)} - \rho_{nv}^{(1)}\mu_{vm}\right].\mathbf{E}(t)
\end{aligned} \tag{2.5.5}$$

In order to evaluate this commutator, the first-order solution is written with changes in the dummy indices as

$$\rho_{vm}^{(1)} = \hbar^{-1}\left(\rho_{mm}^{(0)} - \rho_{vv}^{(0)}\right)\sum_{p}\frac{\mu_{vm} - \mathbf{E}(\omega_p)}{(\omega_{vm} - \omega_p) - i\gamma_{vm}}e^{-i\omega_p t} \tag{2.5.6}$$

and as

$$\rho_{nv}^{(1)} = \hbar^{-1}\left(\rho_{vv}^{(0)} - \rho_{nn}^{(0)}\right)\sum_{p}\frac{\mu_{nv} - \mathbf{E}(\omega_p)}{(\omega_{nv} - \omega_p) - i\gamma_{nv}}e^{-i\omega_p t} \tag{2.5.7}$$

Again the applied optical field $\mathbf{E}(t)$ sum over q is given by

$$\mathbf{E}(t) = \sum_{q}\mathbf{E}(\omega_q)e^{-i\omega_q t} \tag{2.5.8}$$

Inserting Eq. (2.5.7), (2.5.6) and (2.5.8) into Eq. (2.5.5), we have

$$\begin{aligned}\left[\hat{V},\hat{\rho^{(1)}}\right]_{nm} = &-\hbar^{-1}\sum_{v}(\rho_{mm}^{(0)} - \rho_{vv}^{(0)})x\sum_{pq}\frac{[\mu_{nv}.\mathbf{E}(\omega_q)][\mu_{vm}.\mathbf{E}(\omega_p)]}{(\omega_{vm} - \omega_p) - i\gamma_{vm}}e^{-i(\omega_p+\omega_q)t}\\ &+\hbar^{-1}\sum_{v}(\rho_{vv}^{(0)} - \rho_{nn}^{(0)})x\sum_{pq}\frac{[\mu_{nv}.\mathbf{E}(\omega_p)][\mu_{vm}.\mathbf{E}(\omega_q)]}{(\omega_{nv} - \omega_p) - i\gamma_{nv}}e^{-i(\omega_p+\omega_q)t}\end{aligned} \tag{2.5.9}$$

This expression is now inserted into Eq. (2.5.1) and the integration is performed to obtain

$$\begin{aligned}\rho_{nm}^{(2)} = &\sum_{v}\sum_{pq}e^{-i(\omega_p+\omega_q)t}\\ &x\{\frac{\rho_{mm}^{(0)} - \rho_{vv}^{(0)}}{\hbar^2}\frac{[\mu_{nv}.\mathbf{E}(\omega_q)][\mu_{vm}.\mathbf{E}(\omega_p)]}{[(\omega_{nm} - \omega_p - \omega_q) - i\gamma_{nm}][(\omega_{vm} - \omega_p) - i\gamma_{vm}]}\\ &-\frac{\rho_{vv}^{(0)} - \rho_{nn}^{(0)}}{\hbar^2}\frac{[\mu_{nv}.\mathbf{E}(\omega_q)][\mu_{vm}.\mathbf{E}(\omega_q)]}{[(\omega_{nm} - \omega_p - \omega_q) - i\gamma_{nv}][(\omega_{vm} - \omega_p) - i\gamma_{nv}]}\}\\ \equiv &\sum_{v}\sum_{pq}K_{nmv}e^{-i(\omega_p+\omega_q)t}\end{aligned} \tag{2.5.10}$$

We have given the complicated expression in curly braces the label K_{nmv} because appears in many subsequent equations.

We next calculate the expectation value of the atomic dipole moment, which is given by:

$$\langle\vec{\mu}\rangle = \sum_{nm}\rho_{nm}\vec{\mu}_{mn} \tag{2.5.11}$$

We are interested in the various frequency components of $\langle\vec{\mu}\rangle$, whose complex amplitudes $\vec{\mu}(\omega_r)$ are defined through

$$\langle\vec{\mu}\rangle = \sum_r \langle\vec{\mu}(\omega_r)\rangle e^{-i\omega_r t} \tag{2.5.12}$$

Then, in particular, the complex amplitude of the component of the atomic dipole moment oscillating at frequency $\omega_p + \omega_q$ is given by

$$\langle\vec{\mu}(\omega_p + \omega_q)\rangle = \sum_{nmv}\sum_{pq} K_{nmv}\vec{\mu}_{mn} \tag{2.5.13}$$

and consequently the complex amplitude of the component of the nonlinear polarization oscillating at frequency $\omega_p + \omega_q$ is given by

$$\begin{aligned}\mathbf{P}^{(2)}(\omega_p + \omega_q) &= N\langle\vec{\mu}(\omega_p + \omega_q)\rangle \\ &= N\sum_{nmv}\sum_{pq} K_{nmv}\vec{\mu}_{mn}\end{aligned} \tag{2.5.14}$$

Where N denotes the atomic number density. We define the nonlinear susceptibility through the equation

$$\mathbf{P}_i^{(2)}(\omega_p + \omega_q) = \epsilon_0 \sum_{ik}\sum_{pq} \chi^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p)\mathbf{E}_j(\omega_q)\mathbf{E}_k(\omega_p) \tag{2.5.15}$$

By comparison of Eq. (10), (14) and (15), we obtain expression for the second-order susceptibility tensor given by:

$$\begin{aligned}\chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p) &= \frac{N}{\epsilon_0\hbar^2} \\ x\sum_{nmv}\{&(\rho_{mm}^{(0)} - \rho_{vv}^{(0)})\frac{\mu_{mn}^i\mu_{nv}^j\mu_{vm}^k}{[(\omega_{nm} - \omega_p - \omega_q) - i\gamma_{nm}][(\omega_{vm} - \omega_p) - i\gamma_{vm}]} \\ &-(\rho_{vv}^{(0)} - \rho_{nn}^{(0)})\frac{\mu_{mn}^i\mu_{vm}^j\mu_{nv}^k}{[(\omega_{nm} - \omega_p - \omega_q) - i\gamma_{nm}][(\omega_{nv} - \omega_p) - i\gamma_{nv}]}\}\end{aligned} \tag{2.5.16}$$

This is the expression of the second-order susceptibility.

Chapter 3

3. Materials and Method

3.1 Materials

An intensive survey of literature from published articles, books, thesis and dissertation has been carried out on the project.

3.2 Method

3.2.1 Analytical

We have determined the energy eigenvalue of parabolic quantum well by exactly solving the Schrodinger equation using variational technique analytically. Additionally, we have find optical rectification and second-harmonic generation coefficient in a symmetric parabolic quantum well using density matrix formalism.

3.2.2 Computational

The energy eigenvalues of parabolic quantum well were calculated numerically, using MATHEMATICA. We described optical rectification graphically, using MATLAB.

Chapter 4

4. Optical rectification and second order nonlinear optical property

4.1 Introduction

There has been research interest both experimentally and theoretically, in the investigation of low dimensional semiconductor hetrostructure as the result of their intrinsic physical properties and technological applications. The studies on quantum hetrostructures have opened a new field in fundamental physics, and also provide a wide range of potential applications for optoelectronic devices [22]. By changing the profile of a semiconductor quantum well (QW), both the subband energies and their wave functions change, various physical and optical properties depending on them. In this study more attention is given to the second-order nonlinear optical properties such as optical rectification and second-harmonic generation, since the second-order nonlinear optical properties are the lowest-order nonlinear procedures, and the magnitude of the second-order nonlinears are usually stronger than higher-order nonlinearities. In order to obtain a strong second-order optical nonlinearity, the inversion symmetry of the quantum system is broken through applying an electric field to a symmetric parabolic quantum well.

4.2 Mathematical formulation of the problem

We consider a particle with an effective mass of m^* and charge e that moves in the one dimensional potential $V(z) = \frac{1}{2}m^*\omega_0^2 z^2 - eFz$. The first term describes the confining potential of the symmetric parabolic potential well with frequency ω_0 and the second one is the potential energy of the particle in the electric field F. The Schrödinger equation of this problem is

$$\left[\frac{-\hbar^2}{2m^*}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}+\frac{\partial^2}{\partial z^2}\right)+\frac{1}{2}m^*\omega_0^2 z^2 - eFz\right]\psi = E\psi \tag{4.2.1}$$

Where z represents the growth direction of the parabolic quantum well. The eigenfunctions $\psi_{n,k}(x,y,z)$ and the eigenenergies $\epsilon_{n,k}$ are the solutions of Schrödinger equation $\hat{H}\psi_{n,k}(x,y,z) = \epsilon_{n,k}\psi_{n,k}(x,y,z)$ are given by:

$$\begin{aligned}\psi_{n,k}(x,y,z) &= \phi_n(z)u_c(x,y,z)e^{i(k_x x+k_y y)} \text{ and}\\ \epsilon_{n,k} &= E_n + \frac{\hbar^2}{2m^*}\left(k_x^2+k_y^2\right) = E_n + \frac{\hbar^2}{2m^*}|k|^2\end{aligned} \tag{4.2.2}$$

$u_c(x,y,z)$ is the periodic part of the Bloch function in the conduction band at k=0. ϕ_n and E_n are the envelope wave function and the transverse energy of the n^{th} subband. Solutions of the one dimensional Schrödinger equation, $\hat{H}_0\phi_n(z) = E_n\phi_n(z)$, where $\hat{H}_0$ is the z part of the Hamiltonian $\hat{H}$ in Equ. (4.2.1), and given by:

$$\hat{H}_0 = \frac{-\hbar^2}{2m^*}\frac{\partial^2}{\partial z^2} + \frac{1}{2}m^*\omega_0^2 z^2 - eFz \tag{4.2.3}$$

The Schrödinger equation of an electron confined in $\vec{z}$ direction is

$$\left[\frac{-\hbar^2}{2m^*}\frac{\partial^2}{\partial z^2} + \frac{1}{2}m^*\omega_0^2 z^2 - eFz\right]\phi_n(z) = E_n\phi_n(z) \tag{4.2.4}$$

Introducing the dimensionless variable, $\rho = \sqrt{\frac{m^*\omega_0}{\hbar}}\left[z - \frac{eF}{m^*\omega_0^2}\right]$, the spatial co-ordinates, z and dz are described by $z = \sqrt{\frac{\hbar}{m^*\omega_0}}\rho + \frac{eF}{m^*\omega_0^2}$, $dz = \sqrt{\frac{\hbar}{m^*\omega_0}}d\rho$

Equ. (4.2.4) can be rewritten as

$$\left[\frac{-\hbar\omega_0}{2}\frac{d^2}{d\rho^2}+\frac{\hbar\omega_0}{2}\rho^2-\frac{e^2F^2}{2m^*\omega_0^2}\right]\phi_n(\rho)=E_n\phi_n(\rho) \tag{4.2.5}$$

$$\left[\frac{d^2}{d\rho^2}+\frac{2}{\hbar\omega_0}\left(E_n+\frac{e^2F^2}{2m^*\omega_0^2}\right)-\rho^2\right]\phi_n(\rho)=0 \tag{4.2.6}$$

$$\frac{d^2\phi_n}{d\rho^2}+\left(\beta^2-\rho^2\right)\phi_n=0 \tag{4.2.7}$$

Where

$$\beta=\tfrac{2}{\hbar\omega_0}\left(E_n+\tfrac{e^2F^2}{2m^*\omega_0^2}\right)$$

For simplicity let us introduce new variables

$$\delta=\sqrt{\tfrac{m^*\omega_0}{\hbar}}\quad\text{and }\alpha=\tfrac{eF}{m^*\omega_0^2}\text{ transforms }\rho=\delta(z+\alpha)$$

At large ρ, the term β becomes negligible compared to ρ^2 and $\phi_n^{''}\sim\rho^2\phi_n$. This term can be killed by substituting

$$\phi_n(\rho)=\exp\left(\tfrac{-1}{2}\rho^2\right)u(\rho)=u(\rho)e^{\frac{-1}{2}\rho^2}$$

$$\phi_n^{'}=u^{'}(\rho)e^{\frac{-1}{2}\rho^2}+u(\rho)(-\rho)e^{\frac{-1}{2}\rho^2}$$

$$\phi_n^{''}=u^{''}(\rho)e^{\frac{-1}{2}\rho^2}+u^{'}(\rho)(-\rho)e^{\frac{-1}{2}\rho^2}+u^{'}(\rho)(-\rho)e^{\frac{-1}{2}\rho^2}+u(\rho)(-1)e^{\frac{-1}{2}\rho^2}+u(\rho)(\rho^2)e^{\frac{-1}{2}\rho^2}$$

$$=\left[u^{''}(\rho)-2\rho u^{'}(\rho)-u(\rho)+\rho^2u(\rho)\right]e^{\frac{-1}{2}\rho^2} \tag{4.2.8}$$

Substitute Equ. (4.2.8) into Equ (4.2.7), we have

$$u^{''}(\rho)-2\rho u^{'}(\rho)+\rho^2u(\rho)+\left(\beta-\rho^2\right)u(\rho)-u(\rho)=0$$

Finally the differential equation becomes

$$u''(\rho) - 2\rho u'(\rho) + (\beta - 1)u(\rho) = 0 \tag{4.2.9}$$

This ordinary differential equation can be solved by using power series

$$u(\rho) = \sum_{n=0}^{\infty} a_n \rho^n \tag{4.2.10}$$

$$u'(\rho) = \frac{d}{d\rho}\left(\sum_{n=0}^{\infty} a_n \rho^n\right) = \sum_{n=1}^{\infty} a_n n \rho^{n-1} \tag{4.2.11}$$

$$u''(\rho) = \frac{d^2}{d\rho^2}\left(\sum_{n=1}^{\infty} a_n n \rho^{n-1}\right) = \sum_{n=2}^{\infty} a_n n(n-1)\rho^{n-2} \tag{4.2.12}$$

Inserting Equ. (4.2.10),(4.2.11) and (4.2.12) into Equ. (4.2.9), we have

$$\sum_{n=2}^{\infty} a_n n(n-1)\rho^{n-2} - 2\rho \sum_{n=1}^{\infty} a_n n \rho^{n-1} + (\beta - 1)\sum_{n=0}^{\infty} a_n \rho^n = 0$$

By shifting the dummy index of n in the first summation

$$\sum_{n=0}^{\infty} a_{n+2}(n+2)(n+1)\rho^n - 2\sum_{n=1}^{\infty} a_n n \rho^n + (\beta - 1)\sum_{n=0}^{\infty} a_n \rho^n = 0$$

This must hold for all ρ, so the coefficient of each power ρ must be zero. The general result is

$$(n+2)(n+1)a_{n+2} - 2na_n + (\beta - 1)a_n = 0$$

$$a_{n+2} = \frac{2na_n - (\beta - 1)a_n}{(n+2)(n+1)} = \frac{(2n - \beta + 1)a_n}{(n+2)(n+1)} \tag{4.2.13}$$

Equ. (4.2.13) vanishes, when the numerator is equal to zero and the denominator is different from zero.

$$(2n - \beta + 1)a_n = 0$$

$$(2n - \beta + 1) = 0$$

$$\beta = 2n + 1 \tag{4.2.14}$$

But

$$\beta = \frac{2}{\hbar\omega_0}\left(E_n + \frac{e^2F^2}{2m^*\omega_0^2}\right)$$

Thus

$$\frac{2}{\hbar\omega_0}\left(E_n + \frac{e^2F^2}{2m^*\omega_0^2}\right)\text{=2n+1}$$

$$E_n = \frac{1}{2}(2n+1)\hbar\omega_0 - \frac{e^2F^2}{2m^*\omega_0^2}$$

$$E_n = \left(n + \frac{1}{2}\right)\hbar\omega_0 - \frac{e^2F^2}{2m^*\omega_0^2} \tag{4.2.15}$$

The function $u_n(\rho)$ are Hermite polynomials $H_n(\rho)$, and the Hermite function $\phi_n(\rho)$.

$$\phi_n(z) = \left[\frac{\delta}{2^n n!\sqrt{\pi}}\right]^{\frac{1}{2}} \exp\left[\frac{-1}{2}\left(\delta(z+\alpha)\right)^2\right] H_n\left[\delta(z+\alpha)\right] \tag{4.2.16}$$

$$\phi_n(\rho) = \left[\frac{1}{2^n n!\sqrt{\pi}}\right]^{\frac{1}{2}} \exp\left(\frac{-1}{2}\rho^2\right) H_n(\rho) \tag{4.2.17}$$

According to Equ. (4.2.15), the energy eigenvalues and their interval in eV are calculated and shown in table 4.1.

$Fx10^8 V/m$	E_0	E_1	$\triangle E$
0.0	0.26250	0.78750	0.5250
0.3	0.261906	0.786906	0.5250
0.6	0.260124	0.785124	0.5250
0.9	0.257155	0.782155	0.5250
1.1	0.254515	0.779515	0.5250
1.3	0.251348	0.776348	0.5250
1.6	0.245606	0.770606	0.5250
1.9	0.238677	0.763677	0.5250
2.1	0.233398	0.758398	0.5250
2.4	0.224489	0.749489	0.5250
2.7	0.214393	0.739393	0.5250

Table 4.1: *Calculated energy eigenvalue and its interval at confinement frequency of* $8 \times 10^{14}/s$.

From table 4.1, it can be observed that, with the increase of the applied field F, the energy eigenvalues in parabolic quantum well decreases. But, the adjoint energy levels are kept constant.

4.3 Second-order nonlinearity in parabolic quantum well

Recently, nonlinear optical properties in semiconductor quantum wells are of considerable interest because of their relevance in studying practical applications. The lowest-order nonlinearity such as second-order nonlinear optical properties, such as optical rectification, second-harmonic generation and electro-optic effect achieved only for noncentrosymmetric materials. However, centrosymmetric materials, the nonlinear polarization due to three wave mixing process is always zero. The magnitudes of the second-order nonlinears are usually stronger than those of the higher-order nonlinears, if the quantum well has a significant symmetry. In symmetric quantum

well semiconductor structure, the second- order nonlinear susceptibility is zero. Thus, so as to obtain a strong second-order optical nonlinearity, the inversion symmetry of the quantum systems should be broken. These asymmetries can be achieved in two ways; either using advanced material growth technology or by applying an electric field to a symmetric quantum well. In this study, the parabolic quantum well is made asymmetric by applying electric field in the z-direction, and its potential profile is shown in Figure 4.1.

The parameters used are the effective mass of ZnO, $0.21m_0$, confining frequency $\omega_0 = 8x10^{14/s}$ and the applied field $\mathbf{F} = 3x10^{10}V/m$.

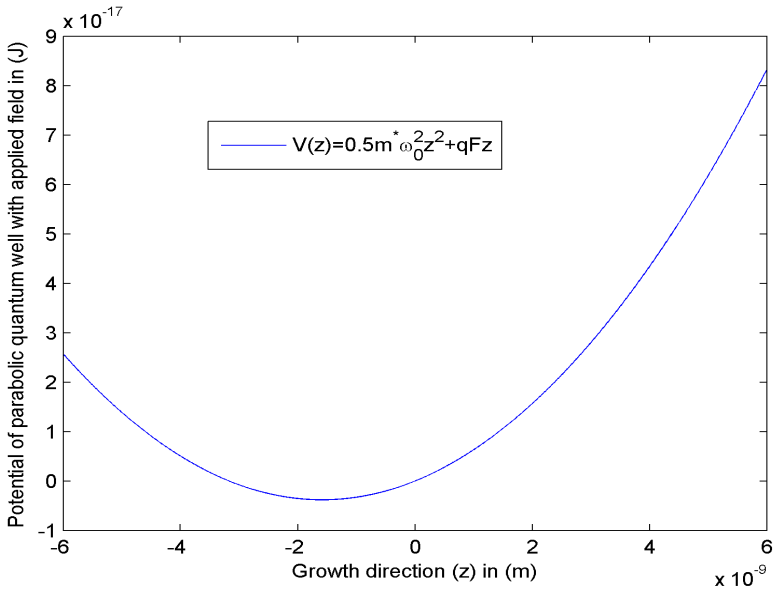

Figure 4.1: *The potential of parabolic quantum well with applied field.*

4.4 Optical rectification

Tera Hertz electromagnetic radiation can be generated via optical rectification of femto second optical pulses incident on an electro-optic material. If an electromagnetic field with a single frequency component $E(t) = Ee^{-i\omega t} + c.c.$, is incidence up on a system with a non zero second-order susceptibility $\chi^{(2)}$, then the nonlinear polarization is

$$\mathbf{P}^2(t) = \epsilon_0\chi^{(2)}\mathbf{E}^2(t) = 2\epsilon_0\chi^{(2)}\mathbf{E}\mathbf{E}^* + \epsilon_0\chi^{(2)}\mathbf{E}^2e^{-i2\omega t} + c.c. \tag{4.4.1}$$

The first term does not oscillate in time. This is known as optical rectification. In this case it produces a static polarization and so does not lead to the generation of radiation(because its second time derivative vanishes). The second term oscillates at frequency 2ω and radiate light at that frequency. This term depends only on the presence of the field at frequency ω and not on the static field. This effect refers to the second-harmonic generation. These and other phenomena arise from a mixing of applied frequencies in a nonlinear medium. Because, the nature of the polarization field is depend on the electric fields. For advanced study of the optical rectification we are interested to use the compact density matrix formalism with applied monochromatic radiation. The evolution of the density matrix after some mathematical manipulation can be given by

$$\dot{\rho}_{nm} = \frac{-i}{\hbar}\left[\hat{H}_0 - \hat{\mu}\mathbf{E}(t), \hat{\rho}\right]_{nm} - \gamma_{nm}\left(\hat{\rho} - \hat{\rho}^{(0)}\right)_{nm} \tag{4.4.2}$$

Where $\hat{H}_0$ is the unperturbed Hamiltonian of the system without considering the applied field, $\hat{\mu}$ is the transition dipole moment, γ_{nm} is the phenomenological damping constant and $\rho^{(0)}$ is the unperturbed density matrix. Equ. (4.4.2) can be solved with

respect to the usual iterative technique [7].

$$\hat{\rho}(t) = \sum_{n} \rho^{(n)}(t) \tag{4.4.3}$$

with

$$\frac{d\rho_{nm}^{(n+1)}}{dt} = \frac{-i}{\hbar}\left\{[\hat{H}_0, \hat{\rho}^{(n+1)}]_{nm} - i\hbar\gamma_{nm}\rho_{nm}^{(n+1)}\right\} + \frac{i}{\hbar}[\hat{\mu}, \hat{\rho}^{(n)}]_{nm}\mathbf{E}(t) \tag{4.4.4}$$

Considering the polarizablity of Equ. (4.4.1), the electron polarization of the n^{th} order is given by

$$\mathbf{P}^n(t) = \frac{1}{A}Tr\left(\hat{\rho}^{(n)}\hat{\mu}\right) \tag{4.4.5}$$

where A is the area of the interaction and the symbol "Tr" denotes the sum of the diagonal elements of the matrix. With the help of the compact density matrix formalism and iterative procedure, the expression for the optical rectification term can be written as

$$\chi_0^{(2)} = \frac{2e^3\alpha_s\mu_{10}^2\delta_{10}}{\epsilon_0\hbar^2}\left[\frac{E_{10}^2}{[(E_{10}-\hbar\omega)^2+(\hbar\gamma)^2]\,[(E_{10}+\hbar\omega)^2+(\hbar\gamma)^2]}\right] \tag{4.4.6}$$

Moreover, the second-harmonic generation term can be given by

$$\chi_{2\omega}^{(2)} = \frac{e^3\alpha_s\mu_{01}\mu_{12}\mu_{20}}{\epsilon_0}\left[\frac{1}{(E_{20}-2\hbar\omega+i\hbar\gamma)\,(E_{10}-\hbar\omega+i\hbar\gamma)}\right] \tag{4.4.7}$$

where α_s is the carrier density, $E_{nm} = E_n - E_m = \hbar\omega_{nm}$ is the energy interval, $\mu_{nm} = \langle\phi_n|z|\phi_m\rangle$ is the off-diagonal matrix element and $\delta_{12} = \mu_{11} - \mu_{00}$, n,m=1,2,3, $E_{10} = E_1 - E_0 = \hbar\omega_{10}$ the energy interval in the parabolic quantum well, and δ_{10} is the mean charge displacement, with $\mu_{10}\delta_{10}$ is the geometric factor of optical rectification. Figure 4.2 describes the optical rectification of the parabolic quantum well versus photon energy with different confining frequency.

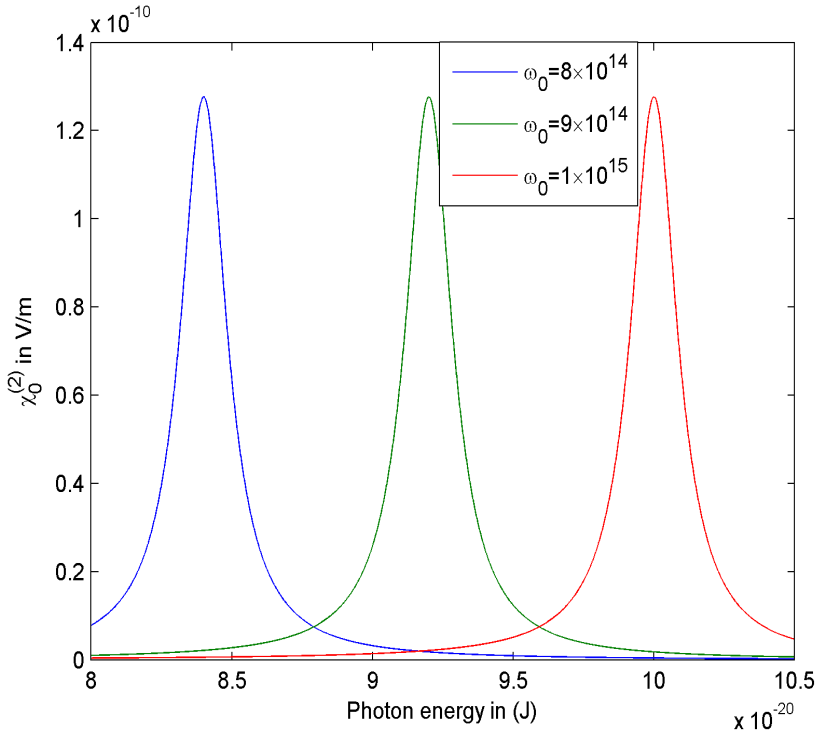

Figure 4.2: *Optical rectification versus photon energy for different confining frequency.*

As it is observed in the Figure, the maxima of optical rectification shifts towards the higher energy, as a confinement frequency of parabolic quantum well increases. The spacing between the energy levels are equal.

In Figure 4.3, we have studied the magnitude of optical rectification for different strengths of electric field. The curves are drawn for three different electric field strengths $F = 0V/m, F = 5x10^7V/m$ and $F = 10x10^7V/m$. The three resonant peaks for parabolic quantum well appears at the same photon energy of 0.525eV. This clearly shows that, for the parabolic quantum well, the adjoint energy levels are kept constant.

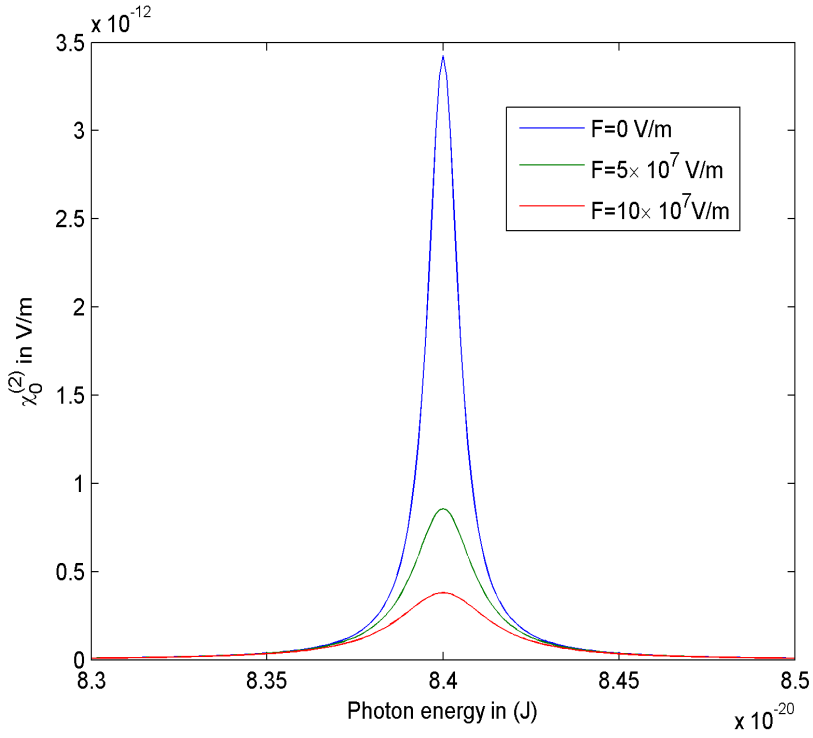

Figure 4.3: *The optical rectification* $\chi_0^{(2)}$ *as a function of photon energy* $\hbar\omega$ *for different strengths of electric field.*

In Figure 4.4, the optical rectification $\chi_0^{(2)}$ as a function of photon energy $\hbar\omega$, for different damping constants $\gamma = 5x10^{11}/s, \gamma = 10x10^{11}/s$ and $\gamma = 15x10^{11}/s$, confining frequency $\omega_0 = 9x10^{14}/s$ and $F = 4x10^7 V/m$, which are shown by blue, green and red lines respectively.

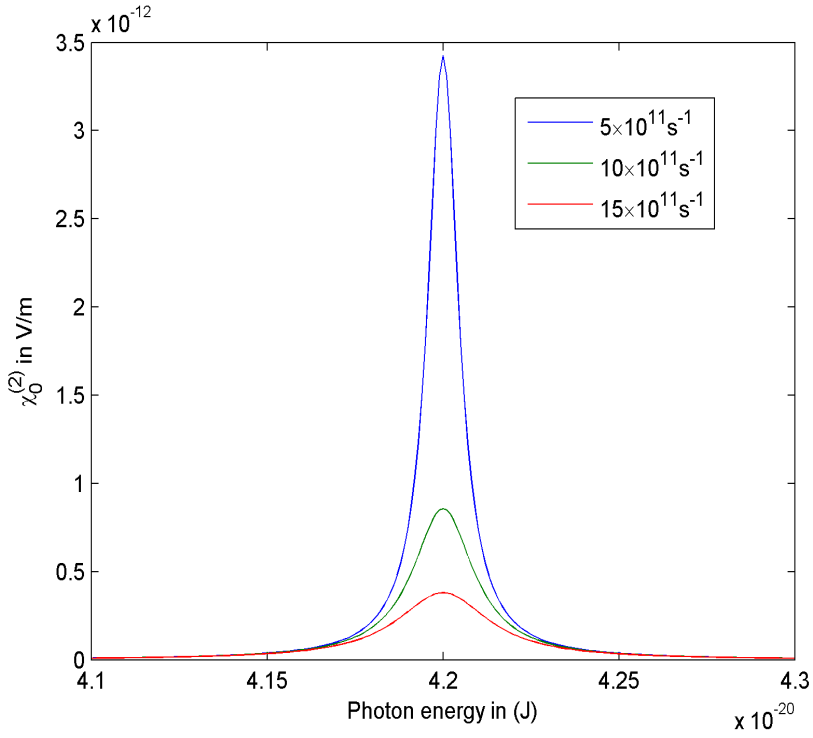

Figure 4.4: *The optical rectification* $\chi_0^{(2)}$ *as a function of photon energy* $\hbar\omega$ *for different phenomenological damping constant.*

From the Figure, it is observed that, the phenomenological damping constant has a great influence on the second-order optical rectification; namely with increase of damping constant, the second-order optical rectification decreases. This is due to electron-electron collision, the electron impurity and electron-phonon scattering interactions. To obtain an amplified second-order susceptibility, it is necessary to reduce these factors.

Chapter 5

Conclusion

In this thesis, we have studied the energy eigenvalue of parabolic quantum well with an applied electric field. The value of ground and first excited state energies are calculated for different value of applied electric field using variational method. Numerical results show that, optical rectification (OR) coefficient is strongly affected by an applied electric field. The energy eigenvalues are decreasing with the increment of an applied electric field. As the magnitude of electric field F increase for parabolic quantum well, the spacing between the energy levels are equal. By using the compact density matrix formalism, the optical rectification (OR) for parabolic quantum well with an applied field has been studied. The optical rectification $\chi_0^{(2)}$ was decreasing with the increment of the magnitude of applied electric field F. Besides, the phenomenological damping constant has a great influence on second-order nonlinear optical rectification (OR). With increase of damping constant, the optical rectification decreases.

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JIMMA UNIVERSITY
COLLEGE OF NATURAL SCIENCES
PERFORMANCE CERTIFICATE FOR MASTER'S DEGREE

Name of Student: **Eliyas Mohammed Jifar** ID No. **SMSC 00906/06**

Graduate Program: **Summer, MSc.**

1. Course Work Performance

Course Code	Course Title	Cr. hr	Number Grade	Rank **	Remark
699	**MSc. Thesis**	**6**	**83.7**	**Very Good**	

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

Thesis Title

Optical rectification and second-order nonlinear optical properties of ZnMgO-ZnO-ZnMgO parabolic quantum wells with applied electric field

2. Board of Examiners decision Mark × in one of the boxes. Pass [×] Failed [ ]
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We the undersigned, number of the Board of Examiners of the final open defense by **Eliyas Mohammed** have read and evaluated his/her thesis entitled "**Optical rectification and second-order nonlinear optical properties of ZnMgO-ZnO-ZnMgO parabolic quantum wells with applied electric field**" and examined the candidate. This is therefore to certify that the thesis has been accepted in partial fulfilment of the requirements for the degree Master of Science in **Physics (Condensed Matter Physics)**.

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DECLARATION

I hereby declare that this Msc dissertation is my original work and has not been presented for a degree in any other University and that all source of materials used for the dissertation have been duly acknowledged.

Name: Eliyas Mohammed Jifar

Signature: _____

email: eliyasmohammad2017gmail.com

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

Name:Dr.Menberu Mengesha

Signature: _____

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