THE RESPONSE OF ELECTRIC FIELD ON NONLINEAR OPTICAL MEDIA

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Abstract

This thesis presents the response of electric field on second order nonlinear optical media. The work focus on purely theoretical study by using anharmonic oscillator model consisting of rectangle shape made of noncentrosymmetric crystal ($LiNbO_3$). The work provides the model equation by employing the assume crystal and using monochromatic laser light on the anharmonic oscillator crystal . In this paper, the formulation of the response functions have been done by applied analytical and computational method on the assumed oscillatory crystal. The thesis provides polarization and susceptibility as a function of frequency which is derived from the model equation. Finally, the paper presents the interaction and the mixing of waves with matter. Moreover, graphical and symbolical interpretations, discussions are involving in the response functions and frequency mixing.

Key Words: Nonlinear responses, Intense field,, Second harmonic generation, Lithium nibonate, Anharmonic oscillator

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Chapter 1

Introduction

Maxwell's theory of electromagnetic field is one of the main pillars of modern theoretical physics and it is playing a key role in the formulation and the development of Einstein's special theory of relativity. The theory of the electromagnetic field, especially in its more advanced stages, has a complex history and many interesting applications. Maxwell's equations are a set of four partial differential equations that to gather with the Lorentz force law and electric circuit[1]. Maxwell's equations represent one of the most elegant and concise ways to state the fundamentals of electricity and magnetism. From those one can develop most of the working relationships in the field. Because of their concise statement, they are embodying a high level of mathematical sophistication.

The basic equations of electricity and magnetism can be used as a starting point for advanced courses, but they are usually first encountered as unifying equations after the study of electricity and magnetic phenomena[1,2]. The Maxwell's equations describe how electric and magnetic fields are generated and altered by each other and by charges and currents. They are named after the physicist and mathematician James clerk Maxwell, who published an early form of those equations between 1861 and 1862. When an electric field is applied to a dielectric material its molecules respond to form microscopic electric dipoles. This Maxwell's equation is also applicable for nonlinear optics[2].

Nonlinear electromagnetic phenomena occur when the response of a medium (including

the electric polarization and its time derivative, the current density, or the magnetization) is a nonlinear function of the applied electric and magnetic field amplitudes. The nonlinearities reside in the constitutive relationships of Maxwell's equations.
Faraday rotation of the plane of polarization of a light wave in an isotropic medium, propagating parallel to an applied magnetic field, could be considered as a nonlinear response in which the optical polarization is a bilinear function of the optical field amplitude and the applied magnetic field. In general, the Maxwell's equations are a key to derive wave function in the interaction of nonlinear media and the optical field. Furthermore,the solution of wave function gives the response functions such as polarization, dielectric, frequency and refractive index [3].

Nonlinear optics is the study of phenomena that occur as the result of the modifications of the optical properties of material system by the presence of light. The beginning of the field of nonlinear optics is often taken to be discovery of second harmonic generation by [8].

Nonlinear optical phenomena are 'nonlinear' in the sense that they occur when the response of a material system to an applied optical field depends on a linear manner on the strength of the electric field, for example the second harmonic generation occurs the 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 applied linear light [20].

Linear and nonlinear response of the medium strongly affects the propagation of electromagnetic waves in the optical materials and can even result in the permanent modification of its physical properties. In turn, the linear and nonlinear optical features of composite materials with metal nanostructures are dominated surface plasma oscillations. The elementary process that is basic in the interaction of light with a medium is the excitation of atoms or molecules by the light field and the remission of light by the excited

particle [4].

The second harmonic generation experiment of Franken et at, marked the birth of the field of nonlinear optics. Harmonic generation of electromagnetic waves at low frequencies had been known for a long time. Harmonic generation of optical waves follow the same principle and should also be observable. The second harmonic generation is the first nonlinear optical effect ever observed in which a coherent input generates a coherent output. But nonlinear optics covers a broader scope. It deals in general with nonlinear interaction of light with matter and includes such problems as light-induced changes of properties of a medium. In general; however, observation of nonlinear optical effects requires the application of lasers.

Numerous nonlinear optical phenomena have been discovered since 1961. They have not only greatly enhanced our knowledge about interaction of light, but also created a revolutionary change in optics technology. Each nonlinear optics process may consist of two parts; the intensity light first induces a nonlinear response in a medium and then medium in reacting modifies the optical fields in a nonlinear away [6]. Two beams of light in the same region of a medium have no effect on each other so that light cannot be used to control light.

The operation of the first laser in 1960 enabled us to examine the behavior of light in optical materials at higher intensities than previously possible.

- The principle of superposition is violated in a nonlinear optical medium.
- The frequency of light is altered as it passes through a nonlinear optical medium;the light can change from red to blue, for example.
- Photons do interact within the confines of a nonlinear optical medium so that light can indeed be used to control light.

The field of nonlinear optics offers a host of fascinating phenomena, many of which are also eminently useful. Nonlinear optical behavior is not observed when light travels in free space. The"nonlinearity" resides in the medium through which the light travels,

rather than in the light itself. The interaction of light with matter is therefore mediated by the nonlinear medium: the presence of an optical field modifies the properties of the medium, which in turn causes another optical field, or even the original field itself, to be modified[11].

1.1 Statement of the Problem

The nonlinear optical properties of materials have been the subject of numerous studies in recent years due to their technological potential applications. Because of their potential uses in nonlinear optical device, the interaction of electric field with nonlinear optical medium shows the wonderful phenomena. The nonlinearities of such media may be strongly enhanced by the electric polarization. The purpose of this work is, therefore, to study the response of the electric field and the modification and generation of new waves using second order nonlinearities, like frequency mixing. Therefore, the this work addresses:

1. How does the nonlinear optics behave?
2. What will be the response of nonlinear media for electric field?
3. How could frequencies mix in second harmonic generation?
4. What would be the important features of in nonlinear optics in media?

The general objective of this study is to understand the optical properties of nonlinear media that responds to the electric field by analytical description of nonlinear polarization, susceptibility, second harmonic generation and frequency mixing. Specifically,the response functions could be derived and interpreted by using the model equations

1.2 Significance of the study

This work will support the development of nonlinear optics in nonlinear media and the improvement of the basic concepts of the nonlinear optics contribution to the electric field;

this also will improve the modification and generation of new waves using second order nonlinearities, like frequency mixing. Nonlinear nature is very important for optical data processing applications and all physics of nonlinear optics can be extracted by studying the behavior of applied optical field-induced polarization of the medium.

1.3 Outlines

The focus of the work presented in this thesis is the study of the response of electric field on nonlinear optical media.

The thesis is organized as follows in five chapters:

The first chapter gives the short introduction to basic properties of the nonlinear optics and the nonlinear crystal . Possible areas of applications as well as the main optical processes are also given. After that, the theoretical concepts of the Maxwell's equations in nonlinear optics are explained and the second harmonic generation also introduces very well in the form of wave mixing or frequency mixing and phase matching.

Chapter 2 gives the brief explanation of the nonlinear optics with formulas. It also contains the derivation of the wave equation by using Maxwell's equation in order to get the solution of nonlinear polarization and the nonlinear susceptibility. The nonlinear functions used in the theoretical calculations of nonlinear optical processing like frequency mixing in second harmonic generation, sum and difference frequency generations.

Chapter 3 presents the analytical and computational methods for formulas of the frequency which is used in this work with description of optical plane-wave by using an harmonic oscillator model.The second harmonic generation is obtained from this model equation by using monochromatic light beam formula. In this chapter, the softwires are used in the thesis like matlab, and inkcape.

In chapter 4,contains the results and discussions,the results are interpreted numerically and graphically. The relation of the electric field, the susceptibility and frequency

are discussed. In general, the response of electric field in nonlinear optical media has been explained and discussed.

The summary ,conclusion and the future work are given in the chapter 5, which completes this work.

Chapter 2

Theoretical Back Ground

Nonlinear optics has been a rapidly growing scientific field in recent decades. It is based on the phenomenon related to the interaction of intense coherent light radiation with matter. The invention of the laser provided enough light intensity that nonlinear optics (NLO) could be observed for the first time. Almost exactly one year after the first ruby laser - second harmonic generation, observed fifty years ago (1961), with a theoretical examination of interactions between light waves in a nonlinear dielectric following very soon thereafter [8,9]. The field has grown so enormously that it is impossible to study all topics.

Optical nonlinearities occur when the output of a material or device ceases to be a linear function of the input power, which is almost always the case for high enough intensities. The nonlinearity may cause a light-induced change in refractive index or absorption of the medium or it may cause new frequencies to be generated.

Nonlinear optics is the study of the interaction of light with matter under conditions in which the nonlinear response of the matter plays an important role. Developments in the field of nonlinear optics hold promise for important applications in optical information processing, telecommunications and integrated optics. Because of the emergence of this field from sold -state physics in which inorganic semiconductors, insulators and crystals have constituted major part of the scientific base.

2.1 Nonlinear Optical Media

The nonlinear terms in the interaction of light with matter give rises to a variety of optical process, in addition to multi -photon absorption. A beam of monochromatic light interaction with matter can be partially converted into light, whose frequencies are harmonic of the fundamental frequency. Nonlinear materials are usually divided into different classes that refer to the order of n of the nonlinear susceptibility χ^n that describes the response of the material to the electric field associated with the light radiation. Nonlinear materials are usually divided into main classes according to the strength of the electric field.
where n is the real number

For instance, χ^2 materials are used for second harmonic generation with the interaction of strong electric field, and χ^3 materials used for the third generation with the interaction of the high intensity electromagnetic field. This work focused on the interaction of nonlinear media with the electric field and the role of polarization (electric dipole moment per unit volume) in nonlinear media. A linear dielectric medium is characterized by a linear relation between the polarization density and the electric field is given by:

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

where: ε_o is the permittivity of free space and $\chi^{(1)}$ is the linear susceptibility of the medium. The relation between polarization and electric field in figure 2.1

A nonlinear dielectric medium, on the other hand, is characterized by a linear relation between Polarization and electric field. The nonlinearity may be microscopic or macroscopic origin. The polarization density is a product of the number of density of dipole moment per volume and the individual dipole moment $\mathbf{p}$ induced by the applied electric field $\mathbf{E}$.

$$\mathbf{P} = N\mathbf{p} \tag{2.1.2}$$

The relation between **P** and **E** is linear when E is small, but becomes nonlinear when E acquires values comparable to intra-atomic electric fields, which are typically 10^5 to $10^8 v/m$ [11]. Since externally applied optical electric fields are typically small in comparison with characteristic interatomic or crystalline fields, the nonlinearity is weak (Changing slightly from linearity, as the electric field increases). The function that relates **P** to **E** can be expanded in a power series[12].

$$\mathbf{P} = \varepsilon_o \chi^{(1)} \mathbf{E} + \varepsilon_o \chi^{(2)} \mathbf{E}^2 + \varepsilon_o \chi^{(3)} \mathbf{E}^3 + ... \tag{2.1.3}$$

where: $\chi^{(1)}$ is the linear susceptibility of the medium which is related to the dielectric constant and the refractive index. The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second and third-order nonlinear optical susceptibilities, respectively.

2.2 Maxwell's Equation In Nonlinear Optics

All electromagnetic phenomena are governed by the Maxwell's equation for the electric and magnetic fields $\mathbf{E}(r,t)$ and $\mathbf{B}(r,t)$ respectively. The four Maxwell's electromagnetic equations are in SI form:

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \tag{2.2.1}$$

$$\nabla \times \mathbf{H} = J + \frac{\partial \mathbf{D}}{\partial t} \tag{2.2.2}$$

$$\nabla \cdot \mathbf{D} = \rho \tag{2.2.3}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{2.2.4}$$

We are primarily interested in the solution of these equations in regions of space that contain no free charge and currents, so that $\rho = 0$ and $\mathbf{J} = 0$. Moreover, we assume that the material is nonmagnetic, thus

$$\mathbf{B} = \mu_O \mathbf{H} \tag{2.2.5}$$

If the material to be nonlinear in the sense that the fields **D** and **E** are related by

$$\mathbf{D} = \varepsilon_O \mathbf{E} + \mathbf{P}_{total} \tag{2.2.6}$$

where in general the polarization vector $\mathbf{P}_{total}$ depends linearly and nonlinearly upon the local value of the electric field strength $\mathbf{E}$. Therefore, the total polarization is the combination of the linear and nonlinear polarization is

$$P_{total} = \mathbf{P}_L + \mathbf{P}_{NL} \tag{2.2.7}$$

Substitute eq.(2.2.8) into eq.(2.2.7), the displacement will be the combination of linear and nonlinear properties. So that

$$\mathbf{D} = \varepsilon_o \mathbf{E} + \varepsilon_o \mathbf{E} + \mathbf{P}_{NL} \tag{2.2.8}$$

$$\mathbf{D} = \varepsilon_o \mathbf{E} + \varepsilon_O \chi \mathbf{E} + \mathbf{P}_{NL} \tag{2.2.9}$$

$$\mathbf{D} = \varepsilon_O (1 + \chi)\mathbf{E} + \mathbf{P}_{NL} \tag{2.2.10}$$

$$D = \varepsilon \mathbf{E} + \mathbf{P}_{NL} \tag{2.2.11}$$

The dielectric function depends on the linear susceptibility as

$$\varepsilon = \varepsilon_O (1 + \chi) \tag{2.2.12}$$

where ε is dielectric constant and $\mathbf{P}_{NL}$ is nonlinear polarization.

We now proceed to derive the optical wave equation in the usual manner. We take the curl of the curl $\mathbf{E}$ Maxwell equation (2.2.1), interchange the order of space and time derivatives on the right-hand side as follows:

$$\nabla \times \nabla \times \mathbf{E} = -\frac{\partial}{\partial t}(\nabla \times \mathbf{B}) \tag{2.2.13}$$

Insert eq (2.2.5) into eq(2.2.13), we will obtain

$$\nabla \times \nabla \times \mathbf{E} = -\frac{\partial}{\partial t}\mu(\nabla \times \mathbf{H}) \tag{2.2.14}$$

Again substitute eq(2.2.2) into eq(2.2.14) and we have

$$\nabla \times \nabla \times \mathbf{E} = -\frac{\partial}{\partial t}\mu(J + \frac{\partial \mathbf{D}}{\partial t}) \tag{2.2.15}$$

Now, let us use Eq. (2.2.11) to eliminate $\mathbf{D}$ from this equation, and we thereby obtain the expression

$$\nabla \times \nabla \times \mathbf{E} = -\mu_O \frac{\partial^2}{\partial t^2}(\varepsilon \mathbf{E} + \mathbf{P_{NL}}) \tag{2.2.16}$$

$$\nabla \times \nabla \times \mathbf{E} = -\mu_O \varepsilon_o \varepsilon \frac{\partial^2}{\partial t^2}\mathbf{E} - \mu_o \frac{\partial^2}{\partial t^2}\mathbf{P_{NL}} \tag{2.2.17}$$

$$\nabla \times \nabla \times \mathbf{E} = -\frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E} - \frac{1}{c^2 \varepsilon_o}\frac{\partial^2}{\partial t^2}\mathbf{P_{NL}} \tag{2.2.18}$$

where μ_o is permeability of free space, $\mu_o = \frac{1}{\varepsilon_o c^2}$ and n is the refractive index,$n = \sqrt{\varepsilon}$. Equation (2.2.18) is the most general form of the wave equation in nonlinear optics. By using an identity, from vector calculus, we can write the first term on the left-hand side of Eq. (2.2.18) as

$$\nabla \times \nabla \times \mathbf{E} = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} \tag{2.2.19}$$

In the linear optics of isotropic source free media, the first term on the right hand side of this equation vanishes because the Maxwell equation $\nabla \cdot \mathbf{D} = 0$ implies that $\nabla \cdot \mathbf{E} = 0$ However, in nonlinear optics this term is generally no vanishing even for isotropic materials, as a consequence of the more general relation (2.2.12) between $\mathbf{D}$ and $\mathbf{E}$ Fortunately, in nonlinear optics the first term on the right-hand side of Eq. (2.2.19) can usually be dropped for cases of interest[7].

If $\mathbf{E}$ is of the form of a transverse, infinite plane wave, $\nabla \cdot \mathbf{E} = 0$ vanishes identically. More generally, the first term can often be shown to be small, even when it does not vanish identically, especially when the slowly varying amplitude approximation is valid. We shall usually assume that the contribution of $\nabla(\nabla \cdot \mathbf{E})$ in Eq.(2.2.19) is negligible so that the wave equation can be taken to have the form

$$\nabla^2 \mathbf{E} = \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E} + \frac{1}{c^2 \varepsilon_o}\frac{\partial^2}{\partial t^2}\mathbf{P_{NL}} \tag{2.2.20}$$

We can interpret this expression as an inhomogeneous wave equation in which the polarization $\mathbf{P}^{NL}$ associated with the nonlinear response drives the electric field $\mathbf{E}$ [10]. This wave equation is needed to be solved to get an expression for nonlinear polarization

and electric field as the function of frequency. The nonlinear wave equation is an inhomogeneous differential equation. The general solution comes from the study of ordinary differential equations and can be solved by the use of a Green's function. Physically one gets the normal electromagnetic wave solution to the homogeneous part of the wave equation is

$$\nabla^2\mathbf{E} = \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E} \tag{2.2.21}$$

Let σ represents wave equation in nonlinear medium.Thus the inhomogeneous equation is given by

$$\sigma = \frac{1}{c^2\varepsilon_o}\frac{\partial^2}{\partial t^2}\mathbf{P_{NL}} \tag{2.2.22}$$

The nonlinear term acts as a driver or sources of electromagnetic waves. One of the consequences of this is a nonlinear interaction that will result in energy being mixed or coupled between different frequencies which is called a wave mixing.

2.3 Second Order Nonlinear Polarization

When light propagates in a transparent medium, its electric field causes some amount of electric polarization in the medium, i.e some density of electric dipole moment. The polarization propagates together with the electromagnetic field in the form of a polarization wave. At low light intensities the electric polarization is proportional to the electric strength, nonlinear contributions become important at high optical intensities laser as light

The second (lowest) order of nonlinear polarization can arise from $\chi^{(2)}$ nonlinearity which can occur only in crystal materials with a non-centrosymmetric crystal structure. The nonlinear polarization has a component which depends quadratically on the electric field of an incident light wave. The nonlinear polarization contains frequency components which are not present in the exciting beam. Light with such frequencies can then be generated in the medium. If the input field is monochromatic, the nonlinear polarization also has a component with twice the input frequency (frequency doubling).

As the polarization has the form of a nonlinear polarization wave, the frequency-doubled light is is also radiated in the direction of the input beam. Other examples are sum and difference frequency generation, optical rectification, parametric amplification and oscillation[33]. Nonlinear optics effects belong to a broader class of electromagnetic phenomena described with in the general frame work of macroscopic Maxwell equations. The Maxwell equations not only serve to identify and classify nonlinear phenomena in terms of the relevant nonlinear optical susceptibilities or nonlinear terms in the induced polarization but also the govern the nonlinear optical propagation effects[10].

We assume the absence of extraneous charges and current, the wave equation is derived in eq(2.2.18). This wave equation is used for representation of nonlinear polarization and nonlinear susceptibility. We represent the polarization in eq(2.2.7). The linear polarization governs linear optical phenomena like optical properties of a medium are independent of the field intensity [4]. 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 in eq.(2.1.1)

In nonlinear optics, the optical response can often be described by generalizing Eq.(2.1.3) by expressing the polarization P(t) as a power series in the field strength E(t). This equation also can be written as

$$\mathbf{P_L}(t) = \mathbf{P^{(1)}}(\mathbf{t}) + \mathbf{P^{(2)}}(\mathbf{t}) + \mathbf{P^{(3)}}(\mathbf{t}) + ... \tag{2.3.1}$$

The quantities $\mathbf{P}^{(2)}$ and $\mathbf{P}^{(3)}$ are known as the second- and third-order nonlinear polarization, respectively. For simplicity, we have taken the fields and polarization to be bold vector quantities in writing Eq.(2.1.3) and eq.(2.3.0). The nonlinear susceptibility is a quantity that is used to determine the nonlinear polarization of a material medium in terms of the strength of an applied optical-frequency electric field.

We shall express $\varepsilon_O \chi^{(2)} \mathbf{E^2}(t)$ as the second-order nonlinear polarization and to $\varepsilon_O \chi^{(3)} \mathbf{E^3}(t)$ as the third-order nonlinear polarization in eq.(2.1.3).

2.4 Second Order Nonlinear susceptibility

The second-order nonlinear susceptibility is an essential parameter for determining the frequency conversion efficiency as well as gain and threshold of such nonlinear-optical devices as harmonic generators and parametric oscillators. It also plays a key role in investigating $\chi^{(2)}$ cascading phenomena which include solitary waves, nonlinear phase shift, and optical processes in second-order nonlinear materials. Knowledge of its absolute values is important for characterizing $\chi^{(3)}$ materials through the $\chi^{(2)}$ cascading process and understanding physics involved in the nonlinear-optical processes.

As a matter of course a large amount of data has been accumulated on the magnitudes of the nonlinear susceptibilities ever since the first observation of second-harmonic generation (SHG) in 1961 [8]. Unfortunately, however, the absolute scale of the second-order nonlinear susceptibilities which should be referred to as standards for various materials and at various wavelengths has not been available; significant discrepancies have been noted among the absolute values reported to date, even for such an important material as $LiNbO_3$.

Nonlinear optical interactions can be described in terms of a nonlinear polarization given by Eq.(2.1.3) only for a material system that is no loss and non dispersion. In the present section, we consider the more general case of a material with dispersion and/or loss. In this more general case the nonlinear susceptibility becomes a complex quantity relating the complex amplitudes of the electric field and polarization. We assume that we can represent the electric field vector of the optical wave as the discrete sum of a number of frequency components as

$$\mathbf{E}(\mathbf{r},\mathbf{t}) = \acute{\sum_n} \mathbf{E_n}(\mathbf{r},\mathbf{t}) \tag{2.4.1}$$

Where

$$\mathbf{E_n}(r,t) = \mathbf{E_n} e^{-i\omega_n t} + c.c \tag{2.4.2}$$

The prime on the summation sign of Eq. (2.4.1) indicates that the summation is to be taken over positive frequencies only. It is also convenient to define the spatially slowly varying field amplitude $\mathbf{E_o}$ by means of the relation

$$\mathbf{E}(r) = \mathbf{E_o} e^{\mathbf{ik_n}} \cdot r \tag{2.4.3}$$

So that

$$\mathbf{E_n}(r,t) = \sum_n \mathbf{E_n} e^{i(\mathbf{k_n}\cdot\mathbf{r}-\omega_\mathbf{n}\mathbf{t})} + c.c \tag{2.4.4}$$

On occasion, we shall express these field amplitudes using the alternative notation $\mathbf{E_n} = \mathbf{E_n}(\omega_n)$ and $\mathbf{E_o} = \mathbf{E_o}(\omega_n)$, $\mathbf{E}(-\omega_n) = \mathbf{E_n}(\omega_n)^*$ and $\mathbf{E_o}(-\omega_n) = \mathbf{E_o}(\omega_n)^*$
Using this new notation, we can write the total field in the more compact form

$$\mathbf{E}(r,t) = \sum_n \mathbf{E}(\omega_\mathbf{n})\mathbf{e}^{-\mathbf{i}\omega_\mathbf{n}\mathbf{t}} = \sum_n \mathbf{E_o}(\omega_\mathbf{n})\mathbf{e}^{\mathbf{i}(\mathbf{k_n}\cdot\mathbf{r}-\omega_\mathbf{n}\mathbf{t}} \tag{2.4.5}$$

where the unprimed summation symbol denotes a summation over all frequencies, both positive and negative. Note that according to our definition of field amplitude, the field given by

$$\mathbf{E}(r,t) = \mathbf{E_o}\cos(\mathbf{k}\cdot r - \omega t) \tag{2.4.6}$$

is represented by the complex field amplitudes

$$\mathbf{E}(\omega) = \frac{1}{2}\mathbf{E_o} e^{ik\cdot r}, \tag{2.4.7}$$

$$\mathbf{E}(-\omega) = \frac{1}{2}\mathbf{E_o} e^{-i\mathbf{k}\cdot r} \tag{2.4.8}$$

or alternatively, by the slowly varying amplitudes

$$\mathbf{E_o}(\omega) = \frac{1}{2}\mathbf{E} \tag{2.4.9}$$

$$\mathbf{E_o}(-\omega) = \frac{1}{2}\mathbf{E} \tag{2.4.10}$$

In either representation, factors of $\frac{1}{2}$ appear because the physical field amplitude $\mathbf{E}$ has been divided equally between the positive and negative frequency field components.

Using a notation similar to that of Eq. (2.4.5), we can express the nonlinear polarization as

$$\mathbf{P}(r,t) = \sum_n \mathbf{P}(\omega_n) e^{-i\omega t} \tag{2.4.11}$$

where, as before, the summation extends over all positive and negative frequency field components.

We now define the components of the second-order susceptibility tensor $\chi^{(2)}_{ijk}(\omega_n + \omega_m, \omega_n, \omega_m)$ as the constants of proportionality relating the amplitude of the nonlinear polarization to the product of field amplitudes according to

$$\mathbf{P}(\omega_n + \omega_m) = \varepsilon_O \sum_{jk} \sum_{nm} \chi^{(2)}_{ijk}(\omega_n + \omega_m, \omega_n, \omega_m) \mathbf{E_j}(\omega_\mathbf{n}) \mathbf{E_k}(\omega_\mathbf{m}) \tag{2.4.12}$$

Here the indices ijk refer to the cartesian components of the fields. The notation (nm) indicates that, in performing the summation over n and m, the sum $\omega_n + \omega_m$ is to be held fixed, although ω_n and ω_m are each allowed to vary. Since the amplitude $\mathbf{E}(\omega_n)$ is associated with the time dependence $exp(-i\omega_n t)$, and the amplitude $\mathbf{E}(\omega_m)$ is associated with the time dependence $exp(-i\omega_m t)$, their product $\mathbf{E}(\omega_n)\mathbf{E}(\omega_m)$ is associated with the time dependence $exp[-i(\omega_n + \omega_m)t]$.

Hence the product $\mathbf{E}(\omega_n)\mathbf{E}(\omega_m)$ does in fact lead to a contribution to the nonlinear polarization oscillating at frequency $\omega_n + \omega_m$, as the notation of Eq. (2.4.8) suggests. Following convention, we have written $\chi^{(2)}$ as a function of three frequency arguments. This is technically unnecessary in that the first argument is always the sum of the other two.

To emphasize this fact, the susceptibility $\chi^{(2)}(\omega_3, \omega_2, \omega_1)$ is sometimes written as $\chi^{(2)}(\omega_3; \omega_2, \omega_1)$ as a reminder that the first argument is different from the other two, or it may be written symbolically as $\chi^{(2)}(\omega_3 = \omega_2 + \omega_1)$.

2.5 Second Order Nonlinear Optics

For two incident waves, one at frequency ω_1 and the other at ω_2 the nonlinear term $\chi^{(2)}$ will introduce (ignoring tensor notation)

$$\mathbf{P}^{(2)} = \chi^{(2)}\mathbf{E}_1 \cos(\omega_1 t - \mathbf{k}_1 z)\mathbf{E_2} cos(\omega^2 t - \mathbf{k}_2 z) \tag{2.5.1}$$

Where amplitudes are $\mathbf{E}_1$ and $\mathbf{E}_2$ and wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are related to frequencies by their respective velocities of light. This product gives two polarization terms, one that oscillates at $\omega_1 + \omega_2$, and the other at $\omega_1 - \omega_2$. Both terms are proportional to the product of the fields.From the quantum mechanical point of view, the nonlinearity has induced two photons to combine into one photon.

When the two photons have the same frequency, one term yields second harmonic and the other yields a term with a static field (the frequency dependence cancels out).

When the incident photons are different, sum and difference frequency photons are generated. An important criterion for materials to exhibit a susceptibility linear in the field, $\chi^{(1)}\mathbf{E}$, is that they contain no center of inversion symmetry. Liquids, gases, amorphous solids and crystalline materials with high symmetry will not directly generate second harmonic[28].

Typically the goal of $\chi^{(2)}\mathbf{E^2}$ terms is to transfer power from one frequency to another, while maintaining a coherent beam. This requires phase-matching, which will be discussed later.

2.5.1 Second Order Nonlinear Optical Processes

In this section we examine the optical properties of a nonlinear medium in which nonlinearities of order higher than the second are negligible, so that

$$\mathbf{P^2}(\mathbf{t}) = \varepsilon_o \chi^2 \mathbf{E^2}(\mathbf{t}) \tag{2.5.2}$$

We consider an electric field $\mathbf{E}$ comprising one or two harmonic components and determine the spectral components of $\mathbf{P^2(t)}$. In accordance with the first Born approximation, the radiation source σ contains the same spectral components as $\mathbf{P^2(t)}$ [11].

Assume that the optical field incident up on the second order nonlinear optical media which consists of two distinct frequency components in the form

$$\mathbf{E(t)} = \mathbf{E_1 e^{-i\omega_1 t}} + \mathbf{E_2 e^{-i\omega_2 t}} + c.c \tag{2.5.3}$$

Then, substitute eq.(2.1.3) into eq(2.5.2), the second-order polarization field in this medium becomes

$$\mathbf{P^{(2)}(t)} = \varepsilon_o \chi^{(2)} [\mathbf{E_1 e^{-i\omega_1 t}} + \mathbf{E_2 e^{-i\omega_2 t}}]^2 \tag{2.5.4}$$

When we expand the squaring part in eq.(2.5.4),we obtain the following frequency components.

$$\mathbf{P^{(2)}(t)} = \varepsilon_o \chi^{(2)} [\mathbf{E_1^2 e^{-2i\omega_1 t}} + \mathbf{E_2^2 e^{-2i\omega_2 t}} + 2\mathbf{E_1 E_2 e^{-i(\omega_1+\omega_2)t}}$$

$$+2\mathbf{E_1 E_2^* e^{-i(\omega_1-\omega_2)t}} + c.c] + \varepsilon_o \chi^{(2)} [\mathbf{E_1 E_1^*} + \mathbf{E_2 E_2^*}] \tag{2.5.5}$$

The resulting polarization field contains components oscillating at difference frequencies. The first term in eq. (2.5.5) oscillates at 2ω and may radiate light at that frequency. This term depends on the presence of the field at frequency ω and not on the static field. This effect is called second harmonic generation (SHG).

Second term oscillates at frequency ω and causes a variation in the refractive index in the medium. This effect is the linear electro-optic effect. The third term does not oscillate in time. This is known as optical rectification.

This is convenient to express this result using the notation

$$\mathbf{P^{(2)}(t)} = \sum \mathbf{P}(\omega_\mathbf{n}) \mathbf{e^{-i\omega_n t}} \tag{2.5.6}$$

where the summation extends over positive and negative frequencies ω_n . The complex amplitudes of the various frequency components of the nonlinear polarization are hence

given by[10].

The two frequency components are second harmonic generation

$$\mathbf{P}(\mathbf{2}\omega_\mathbf{1}) = \varepsilon_O \chi^{(2)} \mathbf{E_1^2} \qquad (\mathbf{SHG}) \tag{2.5.7}$$

$$\mathbf{P}(\mathbf{2}\omega_\mathbf{2}) = \varepsilon_O \chi^{(2)} \mathbf{E_2^2} \qquad (\mathbf{SHG}) \tag{2.5.8}$$

The next frequency component is said to be sum frequency generation

$$\mathbf{P}(\omega_\mathbf{1} + \omega_\mathbf{2}) = 2\varepsilon_O \chi^{(2)} \mathbf{E_1 E_2} \qquad (\mathbf{SFG}) \tag{2.5.9}$$

This component also shows the the difference frequency generation

$$\mathbf{P}(\omega_\mathbf{1} - \omega_\mathbf{2}) = 2\varepsilon_O \chi^{(2)} \mathbf{E_1 E_2^*} \qquad (\mathbf{DFG}) \tag{2.5.10}$$

The last frequency component is said to be optical rectification

$$\mathbf{P}(\mathbf{0}) = 2\varepsilon_O \chi^{(2)} (\mathbf{E_1 E_1^*} + \mathbf{E_2 E_2^*}) \qquad (\mathbf{OP}) \tag{2.5.11}$$

2.5.2 Second Harmonic Generation

Second harmonic generation (SHG) or the frequency doubling generation wave with a doubled frequency(half the wavelength) that two photons are modified and creating a single photon at two times the frequency.

Under proper experimental conditions, the process of second-harmonic generation can be so efficient that nearly all of the power in the incident beam at frequency ω is converted to radiation at the second-harmonic frequency 2ω. One common use of second-harmonic generation is to convert the output of a fixed-frequency laser to a different spectral region.

For example, the Nd:YAG laser operates in the near infrared at a wavelength of $1.06\mu m$. Second-harmonic generation is routinely used to convert the wavelength of the radiation to $0.53\mu m$, in the middle of the visible spectrum.

Second-harmonic generation can be visualized by considering the interaction in terms of the exchange of photons between the various frequency components of the field. According to the picture, which is illustrated in part of Fig. 2.1, two photons of frequency ω

are destroyed, and a photon of frequency 2ω is simultaneously created in a single quantum-mechanical process. The solid line in the figure represents the atomic ground state, and the dashed lines represent what are known as virtual levels. These levels are not energy eigenlevels of the free atom but rather represent the combined energy of one of the energy eigenstates of the atom and of one or more photons of the radiation field[10].

The green laser pointer consists of a diode-pumped solid state laser emitting in the infrared at $1.06\mu m$ that is frequency-doubled by a nonlinear crystal to a wavelength of 0.53nm. For one incident wave of frequency ω_1, the nonlinear term $\chi^{(2)}$ introduces into the polarization $P^{(2)}$ a term that oscillates at $2\omega_1$, the second harmonic.

Applications for SHG go considerably beyond laser pointers. Lasers that directly emit visible light are less efficient than infrared lasers, so when visible light is required, it is preferable to start with the more efficient infrared lasers and to frequency-double them. SHG has been a standard complement for Nd: YAG lasers for a long time. Diode-pumping has replaced lamp-pumping for most of these applications, increasing their efficiency.

Visible (or ultra-violet) lasers are commonly used to pump other lasers, most notably the titanium-sapphire laser (and formerly, the dye laser). The highly inefficient argon laser is rapidly being replaced by frequency-doubled diode-pumped solid state lasers for applications such as pumping the ultra-short pulse titanium-sapphire lasers, which can be mode-locked to pulses only a few femtoseconds long and emit at 800-900 nm wavelengths.

In addition to simply offering light you can see, SHG is important because each visible or UV photon has enough energy to cause a chemical reaction.[25] In non-homogenous materials, the generation of second harmonic may select for specific regions. Examples range from separating out collagen and microtubule in live tissue to observing coupled magnetic and electric domains in ferroelectromagnets [29].

Surface science is an important application because the surface breaks the symmetry of the bulk and enables SHG that depends critically on the character of the surface. The surface can also offer resonance enhancement of the signal [23]; monolayer adsorption can

be detected, for example of tin on GaAs [22, 24]. In other applications, surface SHG can monitor laser melting and separate amorphous from crystalline growth. As a spectroscopic tool, SHG has been used in a plethora of applications, such as probing surface states of metals, surface magnetization, and, using ultra short pulses, a wide range ultrafast surface reactions and surface dynamics.

In this section we present a mathematical description of the process of second harmonic generation, shown symbolically in Fig. 2.1. We assume that the medium is no loss both at the fundamental frequency ω_1 and at the second harmonic frequency $\omega_2 = 2\omega_1$ so that the nonlinear susceptibility obeys the condition of full permutation symmetry. Our discussion closely follows that of one of the first theoretical treatments of second-harmonic generation [9].

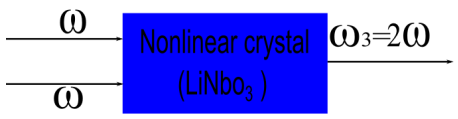

Figure 2.1: Geometry of interaction for second harmonic generation

2.5.3 Sum Frequency Generation

Sum frequency generation (SFG)is the generation of light wave with a frequency that is the sum of two other frequencies. Let us now consider the process of sum-frequency generation, which is illustrated in Fig. 2.2. According to Eq. (2.5.9), the complex amplitude of the nonlinear polarization describing this process is given by the expression

$$\mathbf{P}(\omega_1 + \omega_2) = 2\varepsilon_O \chi^{(2)} \mathbf{E_1} \mathbf{E_2} \tag{2.5.12}$$

In many ways the process of sum-frequency generation is analogous to that of second-harmonic generation, except that in sum-frequency generation the two input waves are at different frequencies. One application of sum-frequency generation is to produce tunable radiation in the ultraviolet spectral region by choosing one of the input waves to be the

output of a fixed-frequency visible laser and the other to be the output of a frequency-tunable visible laser When the incident field E contains two frequencies ω_1 and ω_2 sum frequency generation is possible eq.(2.5.2) the cross-term gives a polarization of the form $\mathbf{P}^{(2)} = \varepsilon_O \chi^{(2)} \mathbf{E_1} \mathbf{E_2}$.

SFG can convert infrared light at frequency ω_2 into a visible signal at frequency $\omega_3 = \omega_1 + \omega_2$. When light at frequency ω_1 is very intense, there is even an effective amplification of the weak infrared signal. Sum Frequency Generation is one way to provide coherent UV light from visible light. If one of the visible lasers has a tunable frequency, the UV light's frequency can be tuned.See figure 2.2

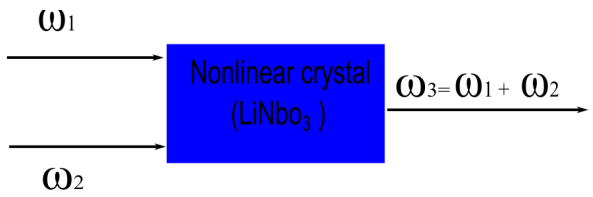

Figure 2.2: Geometry of interaction for sum frequency generation

2.5.4 Difference Frequency Generation(DFG)

Difference frequency generation (DFG) is the generation of light wave with a frequency that is the difference between two other frequencies.

The process of difference-frequency generation is described by a nonlinear polarization of the form

$$\mathbf{P}(\omega_1 + \omega_2) = 2\varepsilon_O \chi^{(2)} \mathbf{E_1} \mathbf{E_2^*} \tag{2.5.13}$$

and is illustrated in Fig. 2.3. Here the frequency of the generated wave is the difference of those of the applied fields. Difference-frequency generation can be used to produce tunable infrared radiation by mixing the output of a frequency-tunable visible laser with

that of a fixed-frequency visible laser.

Difference-frequency generation and sum-frequency generation appear to be very similar processes. However, an important difference between the two processes can be deduced from the description of difference-frequency generation in terms of a photon energy-level diagram of Fig. 2.3.

We see that conservation of energy requires that for every photon that is created at the difference frequency $\omega_3 = \omega_1 - \omega_2$, a photon at the higher input frequency ω_1 must be destroyed and a photon at the lower input frequency ω_2 must be created. Thus, the lower frequency input field is amplified by the process of difference-frequency generation. For this reason, the process of difference-frequency generation is also known as optical parametric amplification.

According to the photon energy-level description of difference-frequency generation, the atom first absorbs a photon of frequency ω_1 and jumps to the highest virtual level. This level decays by a two-photon emission process that is stimulated by the presence of the ω_2 field, which is already present.

Two-photon emission can occur even if the ω_2 field is not applied. The generated fields in such a case are very much weaker, since they are created by spontaneous two photon emission from a virtual level. This process is known as parametric fluorescence and has been observed experimentally [32].

In general, DFG has been used to create infrared light from two higher frequency laser beams. The term DFG usually refers to the case where the beams at the two incident frequencies have comparable intensity. In DFG the interaction of the incoming waves creates a photon of lower energy such that $\omega_3 = \omega_1 - \omega_2$. See figure 2.3
When one beam is very intense and the other is weak, amplification will occur (as with SHG). This is often called parametric amplification, which will be described later.

Difference frequency generation has applications in telecommunications, where wavelength division multiplexing (WDM) puts many wavelengths on the same optical fiber.

In real WDM systems, a way is needed to convert from one wavelength to another.

DFG is attractive in several respects, it is an instantaneous process that can simultaneously convert up and down multiple channels with equal efficiencies, has negligible spontaneous emission noise and no intrinsic frequency chirp [26,27].

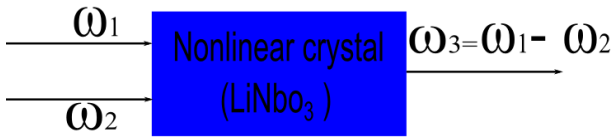

Figure 2.3: Symbolically representation for difference frequency generation

2.5.5 Optical Parametric Oscillation(OPO)

Optical parametric oscillation is the generation of a signal wave and an idler wave by pumping an applied light wave to the resonator.

We have just seen that in the process of difference-frequency generation the presence of radiation at frequency ω_2 and / or ω_3 can stimulate the emission of additional photons at these frequencies. If the nonlinear crystal used in this process is placed inside an optical resonator, as shown in Fig. 2.4, the ω_2 and/or ω_3 fields can build up to large values. Such a device is known as an optical parametric oscillator.

Optical parametric oscillators are frequently used at infrared wavelengths, where other sources of tunable radiation are not readily available. Such a device is tunable because any frequency ω_2 that is smaller than ω_1 can satisfy the condition $\omega_2 + \omega_3 = \omega_1$ for some frequency ω_3.In practice, one controls the output frequency of an optical parametric oscillator by adjusting the phase-matching condition. The applied field frequency ω_1 is often called the pump frequency, the desired output frequency is called the signal frequency, and the other, unwanted, output frequency is called the idler frequency [10].

Figure 2.4: Diagram of optical parametric oscillation

2.5.6 Phase matching

In obtaining efficient SHG, phase-matching is a critical parameter which occurs when the phase velocities of the generated wave and the driving nonlinear polarization wave is matched.[28] Such a phase-matching condition can be obtained when the below vector condition is satisfied:

$$\Delta\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2 = n_1 - n_2 = 0 \tag{2.5.14}$$

However this is difficult to achieve in practice because of the optical dispersion in materials. When there is a significant amount of phase mismatch $\Delta\mathbf{k}$ the SHG intensity exhibits an oscillatory behavior with period $L = \frac{2\pi}{\Delta\mathbf{k}}$ along the crystal. This happens in such a way that after a certain distance which corresponds to half the period, the SHG wave and the driving polarization become 180^0 out of phase and the intensity begins to flow back to the fundamental wave. This distance which equals half the period is called the coherence length:

$$L_c = \frac{\pi}{\Delta\mathbf{k}}$$

In general, for second-order processes we have to satisfy $\omega_1 + \omega_2 = \omega_3$ (energy conservation). For efficient conversion phase-matching is also required, $\mathbf{k_1} + \mathbf{k_2} = \mathbf{k_3}$ (momentum conservation). Since $\mathbf{k} = \omega\frac{n}{c}$, phase-matching is satisfied if $n_1 = n_2 = n_3$. The problem is that materials are usually dispersive and the refractive index varies with frequency.

Normal dispersion has the refractive indices increasing with frequency,$n_3 > n_1, n_2$. This obviously makes it difficult to achieve phase-matching.The most common technique

to obtain phase-matching is to use birefringence to compensate for dispersion. The idea is that at each frequency there is a pair of refractive indices (orthogonal polarizations) and the difference between these is adjusted to compensate for dispersion. The extraordinary refractive index in a uniaxial crystal as a function of angle between the propagation direction and the optic axis is given by[13].

$$n_e(\theta) = (\frac{sin^2\theta}{n_e^2} + \frac{\cos^2\theta}{n_o^2})^{-\frac{1}{2}} \tag{2.5.15}$$

From this, theoretical back ground of the study, we will calculate the response functions to fulfill the gap of the theory by using anharmonic potential energy.

Chapter 3

Methodology

3.1 Formulation of models

Consider an electron which is affected by the deriving force and the damping force. The dynamic motion of electrons shows harmonic and anharmonic oscillation which appears in the restoring force.

The Lorentz model of the atom, which treats the atom as a harmonic oscillator, is known to provide a very good description of the linear optical properties of atomic vapors and of nonmetallic solids. For the case of noncentrosymetric media, like litnium nibanate ($LiNbO_3$) which has good mechamical and chemical stability, the motion of the electron treats as harmonic for linear optics and anharmonic for nonlinearity. In this work, we assume that the applied optical field is given by

$$\mathbf{E(t)} = \mathbf{E_1 e^{-i\omega_1 t}} + \mathbf{E_2 e^{-i\omega_2 t}} \tag{3.1.1}$$

The electric dipole moment in the linear relation will be

$$\mathbf{p} = -e\mathbf{x} \tag{3.1.2}$$

In linear case of N atoms, the polarization vector expresses in the form

$$\mathbf{P(t)} = N\mathbf{p} = -Ne\mathbf{x} \tag{3.1.3}$$

This also can be expressed as in eq.(2.1.1) $\mathbf{P(t)} = \varepsilon_o \chi^{(1)} \mathbf{E(t)}$

In this work, we extend Lorentz model by allowing the possibility of a nonlinearity in

the restoring force exerted on the electron. [10] Here, let us set anharmonic perturbation potential energy in the damping system as shown in figure-3.1.

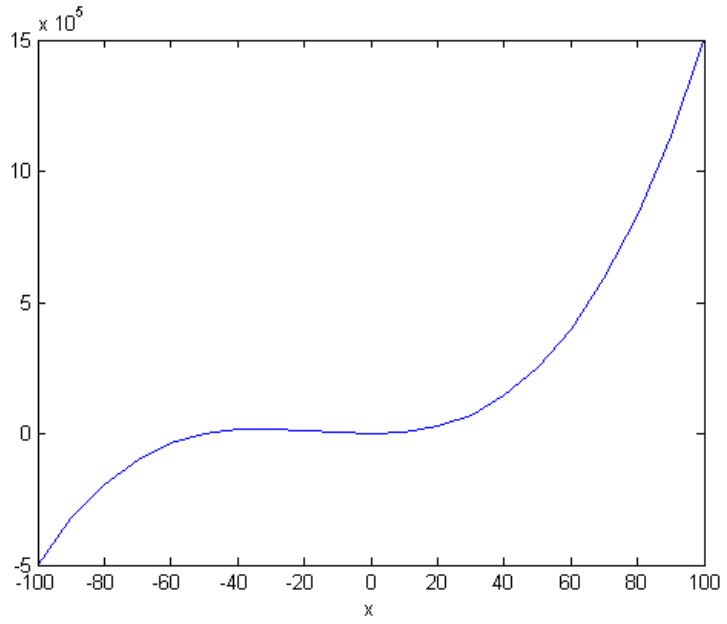

Figure 3.1: Potential energy function for a noncentrosymetric medium k=100N/m,D=3,x= 0 to ±100

$$U(x) = \frac{1}{2}Kx^2 + \frac{1}{3}Dx^3 \tag{3.1.4}$$

The restoring force that corresponding the potential energy function is

$$\mathbf{F_{restoring}}(x) = -\frac{\partial}{\partial x}U(x) = -Kx - Dx^2 \tag{3.1.5}$$

The first term on the right hand side of eq.(3.1.6) represents the linear part and the second hand side of the restoring force denotes the nonlinearity.

where D is a parameter that characterizes the strength of the nonlinearity.

Therefore, we derived the restoring force which shows the nonlinear function of the the restoring force acting on the electron and retaining the linear and the quadratic terms in the Taylor series expansion of the restoring force in the displacement(x).

The forces that acting on the dynamics motion of the electron are the restoring force

$F_{restoring}$, the applied (deriving)force F_e and the damping force F_d

The damping force is proportional to the velocity of the electron. Thus

$$\mathbf{F}_d = -b\dot{x} \tag{3.1.6}$$

where $b = 2\gamma m$ and γ is the damping parameter

The driving force is given by

$$\mathbf{F} = -e\mathbf{E}(\mathbf{t}) \tag{3.1.7}$$

where $\mathbf{E}(\mathbf{t}) = \mathbf{E_1}e^{-i\omega_1 t} + \mathbf{E_2}e^{-i\omega_2 t}$

The equation of anharmonic oscillation of the electron is of the form

$$\ddot{x} + 2\gamma\dot{x} + \omega_o^2 x + Dx^2 = -\frac{e}{m}\mathbf{E}(\mathbf{t}) \tag{3.1.8}$$

No general solution to eq.(3.1.9) for an applied field of the form eq.(3.1.1), if the applied field is sufficiently weak, then the nonlinear term Dx^2 will be much smaller than the linear term $\omega_o^2 x$ for any displacement x that can be induced by the field.

Under this condition eq.(3.1.9) can be solved by means of a perturbation expansion. Let us substitute $\mathbf{E}(\mathbf{t})$ into eq.(3.1.9) by $\lambda\mathbf{E}(\mathbf{t})$

where λ is a perturbation that ranges continuously between zero and one, and characterizes the strength of the perturbation that will be set equal to one at the end of the calculation. Thus it becomes

$$\ddot{x} + 2\gamma\dot{x} + \omega_o^2 x + Dx^2 = -\lambda\frac{e}{m}\mathbf{E}(\mathbf{t}) \tag{3.1.9}$$

We should find the solution to eq.(3.1.10) in the form of a power series expansion in the strength λ of the perturbation, that is, the solution of the form

$$x = \lambda^1 x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + ... \tag{3.1.10}$$

Insert eq(3.1.11) in to eq.(3.1.10) for value of the coupling strength λ, the term in this equation is proportional to $\lambda^1, \lambda^2, \lambda^3$, etc, each of them satisfy the equation separately.

Now, express eq.(3.1.9) as follows

$$\frac{\partial^2 x}{\partial t^2} + 2\gamma\frac{\partial x}{\partial t} + \omega_o^2 x + Dx^2 = -e\frac{E(t)}{m} \tag{3.1.11}$$

and substitute eq.(3.1.11)into eq.(3.1.12)

$\frac{\partial^2[\lambda^1 x^{(1)}+\lambda^2 x^{(2)}+\lambda^3 x^{(3)}+...]}{\partial t^2} + 2\gamma\frac{\partial[\lambda^1 x^{(1)}+\lambda^2 x^{(2)}+\lambda^3 x^{(3)}+...]}{\partial t}+$

$$\omega_o^2[\lambda^1 x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + ...] + D[\lambda^1 x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + ...]^2 = -e\frac{E(t)}{m} \tag{3.1.12}$$

From eq.(3.1.13), we have separate terms, these terms leads to the following equations respectively.

The first order,

$$\ddot{x}^{(1)} + 2\gamma\dot{x}^{(1)} + \omega_o^2 x^{(1)} = -e\frac{E(t)}{m} \tag{3.1.13}$$

The second order,

$$\ddot{x}^{(2)} + 2\gamma\dot{x}^{(2)} + \omega_o^2 x^{(2)} + D[x^{(1)}]^2 = 0 \tag{3.1.14}$$

The thrid order,

$$\ddot{x}^{(3)} + 2\gamma\dot{x}^{(3)} + \omega_o^2 x^{(3)} + 2Dx^{(1)}x^{(2)} = 0, etc \tag{3.1.15}$$

Now, neglecting the higher orders $x^{(3)}$ and the lowest order contribution $x^{(1)}$ is governed by the same equation as the result of the linear Lorenz model. Thus, the solution of the linear equation is given by

$$x^1(t) = x^{(1)}(\omega_1)e^{-i\omega_1 t} + x^{(1)}(\omega_2)e^{-i\omega_2 t} + c.c \tag{3.1.16}$$

From this eq.(3.1.17) we calculate $\frac{\partial x}{\partial t}$ and $\frac{\partial^2 x}{\partial t^2}$ to obtain the amplitude $x^{(1)}(\omega_j)$ in the form

$$x^{(1)}(\omega_j) = -\frac{e}{m}(\frac{E_j}{\omega_o^2 - \omega_j^2 - 2i\omega_j\gamma}) \tag{3.1.17}$$

Let us introduce the the complex denominator function

$$G(\omega_j) = \omega_o^2 - \omega_j^2 - 2i\gamma\omega_j \tag{3.1.18}$$

Now, put the complex denominator into eq(3.1.18) and we have

$$x^{(1)}(\omega_j) = -\frac{e}{m}\frac{E_j}{G(\omega_j)} \tag{3.1.19}$$

To find the linear susceptibility, we should relate the dipole moment and the polarization as the function of frequency. The dipole moment as the function of frequency will be

$$\mathbf{p}(\omega_{\mathbf{j}}) = -ex^{(1)}(\omega_j) \tag{3.1.20}$$

For N-density of atoms, the linear contribution to the polarization is given by

$$\mathbf{P}(\omega_j) = \varepsilon_o \chi^{(1)} \mathbf{E}(\omega_j) \tag{3.1.21}$$

$$\mathbf{P}(\omega_j) = N\mathbf{p}_{\mathbf{j}} = -eNx^{(1)}(\omega_j) \tag{3.1.22}$$

We solve the linear susceptibility by combining eq.(3.1.22) and (3.1.23)

$$\chi^{(1)}(\omega_j) = \frac{Ne^2}{\varepsilon_o m G(\omega_j)} \tag{3.1.23}$$

Let us find the response function in nonlinearity from eq.(3.1.15). Substitute eq.(3.1.20) into eq.(3.1.15) and we have the following

$$\ddot{x}^{(2)} + 2\gamma\dot{x}^{(2)} + \omega_o^2 x^{(2)} = D[-\frac{e}{m}\frac{E_j}{G(\omega_j)}]^2 \tag{3.1.24}$$

We should solve the following equation to find the response at frequency

$$\ddot{x}^{(2)} + 2\gamma\dot{x}^{(2)} + \omega_o^2 x^{(2)} = D[-\frac{e}{m}\frac{E_1}{G(\omega_j)}]^2 e^{2i\omega_1 t} \tag{3.1.25}$$

Therefore, the solution of this equation will be

$$x^{(2)}(t) = x^{(2)}(2\omega_1)e^{2i\omega_1 t} \tag{3.1.26}$$

From eq.(3.1.27), find the following expression $\frac{\partial^2 \ddot{X}^{(2)}}{t^{(2)}}$ and $\frac{\partial \dot{X}^{(2)}}{\partial t}$ and substitute into eq.(3.1.26) we have this results

$$x^{(2)}(2\omega_1) = -\frac{D(\frac{e}{m})^2 E_1^2}{G(2\omega_1)G^2(\omega_1)} \tag{3.1.27}$$

In the same manner, the amplitude of the response at the other frequencies are found to be

$$x^{(2)}(2\omega_2) = \frac{-D(\frac{e}{m})^2 E_2^2}{G(2\omega_2)G^2(\omega_2)} \tag{3.1.28}$$

At frequency $(\omega_1 + \omega_2)$, we will have

$$x^{(2)}(\omega_1 + \omega_2) = \frac{-2D(\frac{e}{m})^2 E_1 E_2}{G(\omega_1 + \omega_2)G(\omega_1)G(\omega_2)} \tag{3.1.29}$$

At frequency $(\omega_1 - \omega_2)$, the expression will be

$$x^{(2)}(\omega_1 - \omega_2) = \frac{-2D(\frac{e}{m})^2 E_1 E_2}{G(\omega_1 - \omega_2)G(\omega_1)G(-\omega_2)} \tag{3.1.30}$$

At frequency $(\omega_1 - \omega_1) = (\omega_2 - \omega_2) = 0$, it becomes

$$x^{(2)}(0) = \frac{-2D(\frac{e}{m})^2 E_1 E_2^*}{G(0)G(\omega_1)G(-\omega_1)} + \frac{-2D(\frac{e}{m})^2 E_1 E_2^*}{G(0)G(\omega_2)G(-\omega_2)} \tag{3.1.31}$$

After this result, nonlinear susceptibility can be calculated as an analogous to the linear expression. So that the nonlinear susceptibility in second harmonic generation will be

$$\mathbf{P}^{(2)}(2\omega_1) = \varepsilon_0 \chi^{(2)}(2\omega_1, \omega_1, \omega_1) E^2(\omega_1) \tag{3.1.32}$$

where $P^{(2)}(\omega_1)$ is the amplitude of the component of the nonlinear polarization oscillating at frequency $2\omega_1$ and is defined by the relation

$$\mathbf{P}^{(2)}(2\omega_1) = -Nex^{(2)}(2\omega_1) \tag{3.1.33}$$

By equating eq(3.1.33) and (3.1.34), we have the following expression

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{N(\frac{e^3}{m^2})D}{\varepsilon_O G(2\omega_1)G^2(\omega_1)} \tag{3.1.34}$$

Using eq(3.1.24), the result can be written in terms of product of linear susceptibilities like.

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{\varepsilon_o^2 mD}{N^2 e^3} \chi^{(1)}(2\omega_1)[\chi^{(1)}(\omega_1)]^2 \tag{3.1.35}$$

The nonlinear susceptibility for second-harmonic generation of the ω_2 field is obtained trivially from Eq.(3.1.35) and (3.1.36) through the substitution $\omega_1 \to \omega_2$. The nonlinear susceptibility describing sum-frequency generation is obtained from the relations

$$\mathbf{P}(\omega_1 + \omega_2) = 2\varepsilon_O \chi^{(2)}(\omega_1 + \omega_2, \omega_1, \omega_2)\mathbf{E}(\omega_\mathbf{1})\mathbf{E}(\omega_\mathbf{2}) \tag{3.1.36}$$

and

$$\mathbf{P}^{(2)}(\omega_1+\omega_2) = -Ne\chi^{(2)}(\omega_1+\omega_2) \tag{3.1.37}$$

Note that in this case the relation defining the nonlinear susceptibility contains a factor of two because the two input fields are distinct, as discussed in relation to Eq. (3.1.34). By comparison of these equations with (3.1.30), the nonlinear susceptibility is seen to be given by

$$\chi^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2) = \frac{N(\frac{e^3}{m^2})D}{\varepsilon_O G(\omega_1+\omega_2)G(\omega_1)G(\omega_2)} \tag{3.1.38}$$

. This result can be expressed in terms of the product of linear susceptibilities as

$$\chi^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2) = \frac{\varepsilon_o^2 mD}{N^2e^3}\chi^{(1)}(\omega_1+\omega_2)\chi^{(1)}(\omega_1)\chi^{(1)}(\omega_2) \tag{3.1.39}$$

The nonlinear susceptibilities describing the other second-order processes are obtained in an analogous manner.

For difference-frequency generation, we have

$$\chi^{(2)}(\omega_1-\omega_2,\omega_1,-\omega_2) = \frac{N(\frac{e^3}{m^2})D}{\varepsilon_O G(\omega_1-\omega_2)G(\omega_1)G(-\omega_2)}$$

$$= \frac{\varepsilon_o^2 mD}{N^2e^3}\chi^{(1)}(\omega_1-\omega_2)\chi^{(1)}(\omega_1)\chi^{(1)}(-\omega_2) \tag{3.1.40}$$

For optical rectification, the nonlinear susceptibility will be

$$\chi^{(2)}(0,\omega_1,-\omega_2) = \frac{N(\frac{e^3}{m^2})D}{\varepsilon_O G(\omega_1-\omega_2)G(\omega_1)G(-\omega_2)}$$

$$= \frac{\varepsilon_o^2 mD}{N^2e^3}\chi^{(1)}(0)\chi^{(1)}(\omega_1)\chi^{(1)}(-\omega_2) \tag{3.1.41}$$

The analysis just presented shows that the lowest-order nonlinear contribution to the polarization of a noncentrosymmetric material is second order in the applied field strength. This analysis can readily be extended to include higher-order effects. The solution to Eq. (3.1.16), for example, leads to a third order or $\chi^{(3)}$ susceptibility, and more generally terms proportional to λ^n the expansion described by Eq. (3.1.11) lead to a $\chi^{(n)}$ susceptibility.

3.2 Method and Study design

We used analytical and computational methods to study the nonlinear second order susceptibility, electric polarization and frequency mixing in nonlinear second order optics.

3.2.1 Analytical Method

In this work, the most important method deriving equations analytically is very essential input for numerical computation.

3.2.2 Computational metthod

In this study we started from the power series representation of the polarization in terms of applied field. The electric field is also as the function of time and frequency in the second harmonic-Generation and we calculated the value of nonlinear optical susceptibility and mixing frequency in different mechanisms that leads to optical nonlinearities

3.3 Methods and softwares used

The study is purely theoretical understanding of the optical properties of the nonlinear media and the interaction of optical field and the media.

Softwares are the programs that run on a computer and perform certain functions. These materials are essential to have good understanding and manipulating, interpreting numerically and graphically the results of the work.

We employed computational methods using Matlab program by developing suitable computer codes for studying the response functions and the properties of nonlinear optical media by applying monochromatic optical field. We also developed inkscape program to draw or plot figures

In the study we started from model equation that describes the responses as a function of electric field. we employed Taylor expansion that related the polarization, the susceptibility and the electric field.

After we have derived the model equation that the responses as a function of frequency, we carried out Matlab program for our model equation to generate data and plot the graphs in different legends.

Chapter 4

Results and Discussions

4.1 Lithium nibonate as nonlinear crystal

We now interpret the results of the formulation of the response functions in assumed model. We took Lithium niobate ($LiNbO_3$) for our model crystal as second nonlinear material. This material is well understood, that has a good mechanical and chemical stability and sufficiently large nonlinear optical coefficient, that is why this crystal is taken as model.

4.2 Nonlinear response functions

To this end, by considering a monochromatic input field in the assuming model

$$\mathbf{E}(t) = \mathbf{E}_1 e^{-i\omega_1 t} + \mathbf{E}_2 e^{-i\omega_2} t + c.c$$

and this model can be expressed using anharmonic potential.

$$\ddot{x} + 2\gamma\dot{x} + \omega_o^2 x + Dx^2 = -\frac{e}{m}\mathbf{E}(\mathbf{t})$$

Based on our model equation, we calculated the following response functions, such as linear and nonlinear polarization, linear and nonlinear susceptibility and frequency mixing

4.3 Polarization in nonlinear media

From formulation of the model, the linear and nonlinear polarization are derived as

$\mathbf{P}(\omega) = \varepsilon_O \chi^{(1)} \mathbf{E}(\omega)$ and $\mathbf{P}(\omega) = \varepsilon_O \chi^{(2)} \mathbf{E}^2(\omega)$ respectively.

The following graphs shows the relation between the polarization and the electric field

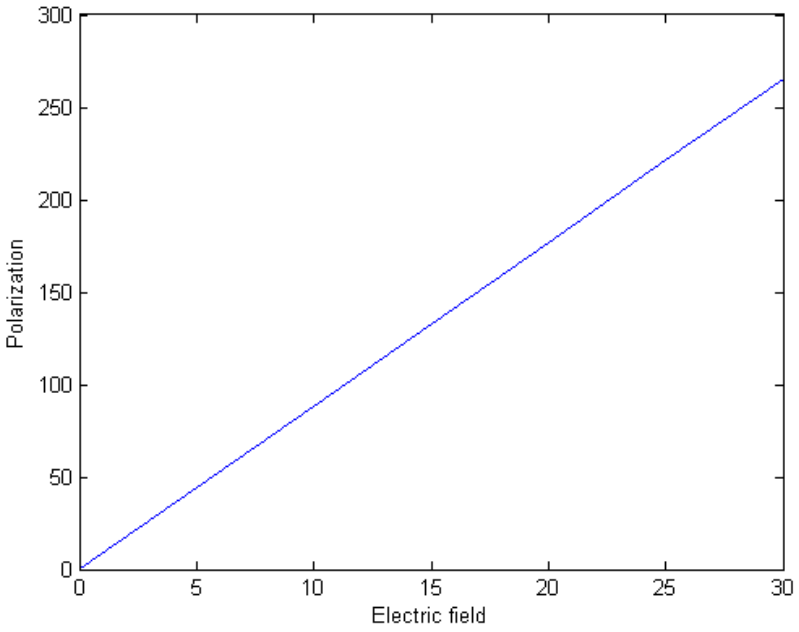

Figure 4.1: Linear polarization versus electric field in linear medium,taking as $\varepsilon_o = 8.85 \times 10^{-12} F/m$, $\mathbf{E} = 0 - 30v/m$

According to [10] approximation, $\chi^{(2)} = 6.9 \times^{-12} m/v$ and ε_o =$8.85 \times 10^{-12} F/m$, $\mathbf{E} = (0 - 10^{10})v/m$, $d = \varepsilon_o \chi^{(2)} = 4.213485 \times 10^{-34}$

In fig.4.1, the polarization $\mathbf{P_L}$ is linearly proportional to the electric field in linear materials or at low optical field where as fig.4.2, indicated that the relation between nonlinear polarization $\mathbf{P_{NL}}$ and applied electric field is quadratic in nonlinear optical media at strong field, that is:

$\mathbf{p}_{NL}(\omega) = \varepsilon_o \chi^{(2)} \mathbf{E}^2(\omega)$

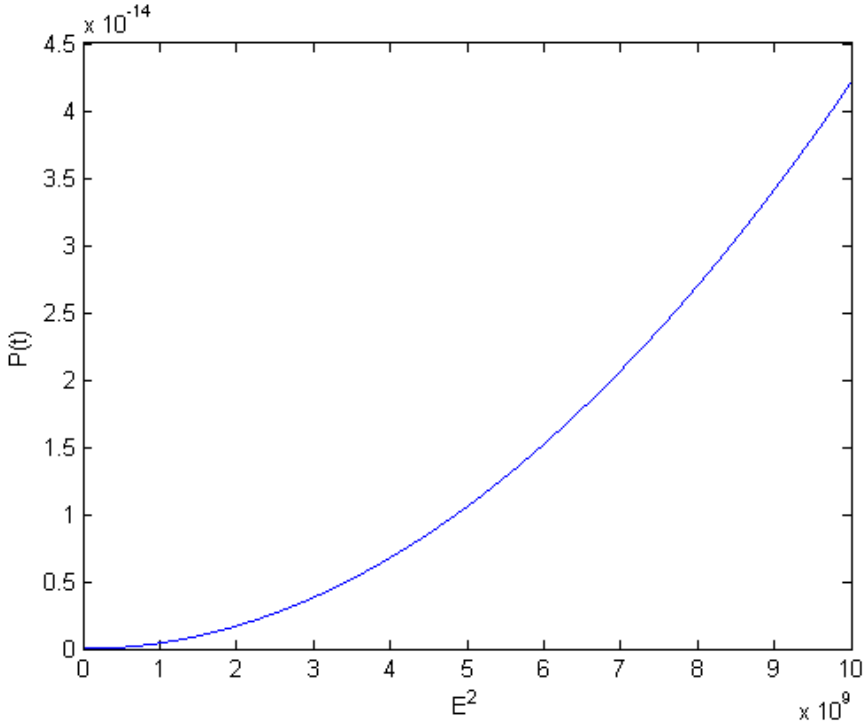

Figure 4.2: Nonlinear polarization versus electric field in nonlinear medium

4.4 Susceptibility in nonlinear media

According to model equation, the linear and nonlinear susceptibilities also has been obtained

$$\chi^{(1)}(\omega_j) = \frac{Ne^2}{\varepsilon_o m G(\omega_j)} \text{ and } \chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{N(\frac{e^3}{m^2})D}{\varepsilon_o G(2\omega_1) G^2(\omega_1)}$$

The susceptibility is proportional to the polarization linearly at low applied field where as the susceptibility $\chi^{(2)}$ is proportional to the nonlinear polarization quadratically. This means, as the electric field increases, the electric susceptibility also increases in nonlinear media.

4.5 Frequency mixing in nonlinear medium

For a second order nonlinear process involving the interaction of three optical frequencies $\omega_1, \omega_2, \omega_3$, the waves can be expressed as energy diagram in the next subsection.

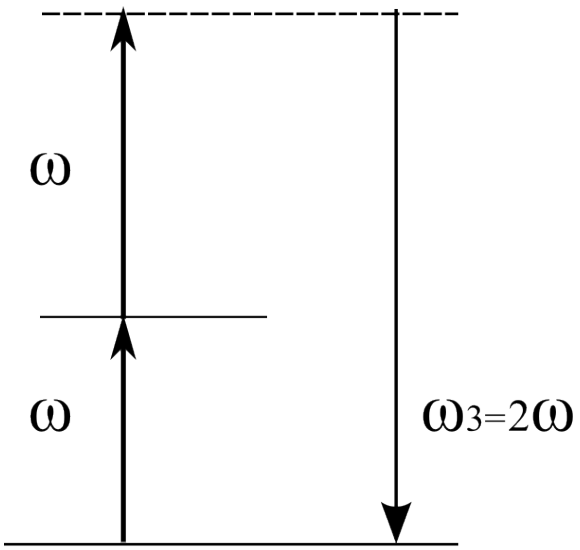

Figure 4.3: Diagram of energy level for SHG

In this energy diagram, two the same-frequency photons are absorbed and double photon with same frequency was created.

The photon energy diagram for sum frequency generation described in fig.4.4. as

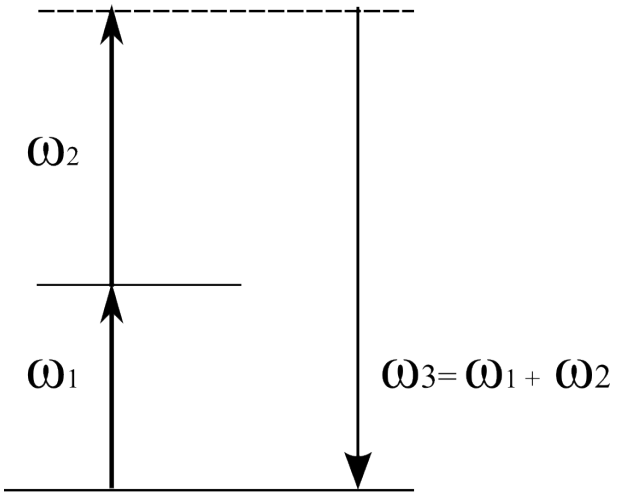

Figure 4.4: Diagram of photon energy level for SFG

In this energy level, the diagram indicates. two low-frequency photons are absorbed and one photon with high frequency was created. This is, $\omega_3 = \omega_1 + \omega_2$

Energy level of photons for difference frequency generation in fig.4.5. are

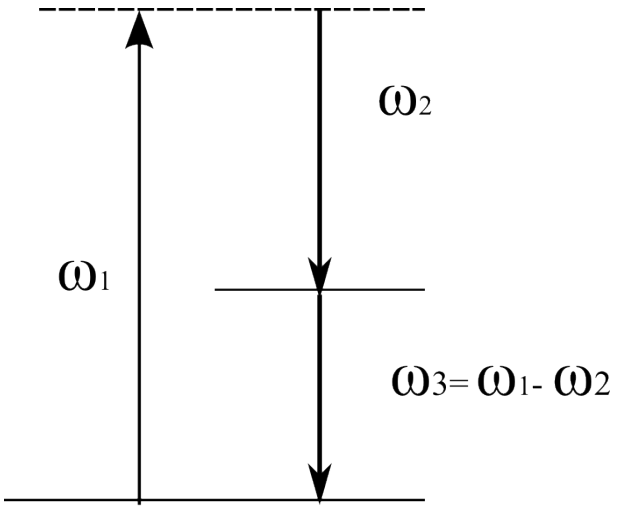

Figure 4.5: Diagram of energy level for DFG

The two incoming waves are interacting, one which has high frequency is absorbed and another is created, finally, the signal wave is $\omega_3 = \omega_1 - \omega_2$

Photon Energy level diagram for optical parametric oscillation in fig.4.6. are shown.

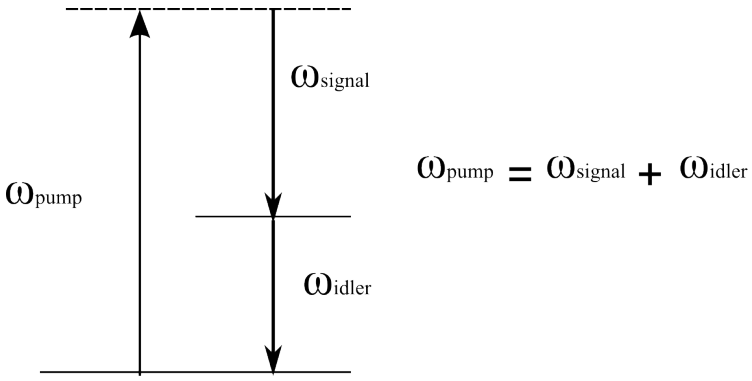

Figure 4.6: Diagram of energy level for OPO

In figure 4.6, annihilation of one high frequency photon and creation of two low-frequency photons expressed as , $\omega_1 = \omega_2 + \omega_3$

Chapter 5

Conclusion and Future outlook

Considering Litinium Nibanate ($LiNbo_3$) as assumed anharmonic oscillatory model nonlinear crystal and employing the model equation by using analytical and computational method, we obtained expressions of the response functions like nonlinear polarization and susceptibility as the function of frequency. From anharmonic oscillatory model we understood that when an electromagnetic wave propagates through nonlinear medium, an electric field acts on each particle displacing them from their equilibrium positions, In such a case, positively charged cores and surrounding negatively charged electrons are displaced in opposite directions. This leads to the separation of opposite charges, creating dipoles in the medium.

From this, we concluded that the response functions are ultimately due to inability of dipoles in the nonlinear optical medium. In this thesis, we observed that the relation between the second order nonlinear polarization and the electric field is quadratic. Therefore, we concluded that, as the electric field intensity increases, the nonlinear polarization and nonlinear susceptibility also increase in the nonlinear media. That is, at such high intensity fields, the optical response of the medium to the radiation becomes nonlinear. In addition to this,the optical properties of the nonlinear medium depend on the applied field. Moreover, the interaction of the field and matter in the model crystal will absorb or create photons.

In our calculation, we observed that the optical processes such as second harmonic generation (SHG), sum frequency generation (SFG), difference frequency generation (DFG) are the maim results. In general, we concluded that frequency mixing in second order nonlinear medium and the response functions are due to the nonlinearity .

For future work we can study the properties of nonlinear optics in optical medium specially on our assuming model ($LiNbO_3$) in order to understand the the responses and the properties of the $LiNbO_3$ nonlinear crystals in experimentally.

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