DETERMINATION CHANGE IN THIRD ORDER NONLINEAR REFRACTIVE INDEX AND ABSORPTION COEFFICIENT OF GALLIUM ARSENIDE USING NONLINEAR ELECTRON OSCILLATOR MODEL

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This is to certify that the thesis prepared by **Tigist Woldeyes** Graduate Studies entitled "**Determination change in third order nonlinear refractive index and absorption Coefficient of Gallium Arsenide using nonlinear electron oscillator model** " in fulfillment of the requirements for the degree of complies with the regulations of the University and meets the accepted standards with respect to originality and quality. .

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To my family.

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Abstract

The optical properties of a semiconductor can be defined as any property that involves the interaction between electromagnetic radiation or light and the semiconductor, including absorption, diffraction, polarization, reflection, refraction, and scattering effects.

The main aim of this research work is to understand the nonlinear optical properties of GaAs. In doing so, the third order nonlinear absorption coefficient and refractive index will be analyzed numerically using anharmonic oscillator model.In the Lorentz model the motion of the electrons in the medium is treated as a harmonic oscillator. This can be pictured as electrons attached to their nuclei by springs with resonant frequency ω and dampingγ.

An optical wave provides a forcing term through the dipole interaction with the electron and hence the motion of the electron around its equilibrium position can be described by the linear differential equation, Analytical and numerical studies have been made to describe the dependence of third order nonlinear susceptibility on laser frequency and vibrational frequency Gallium Arsenide (GaAs).

The frequency response of the real and imaginary part of third order nonlinearity is studied using anharmonic oscillator model. The change in real and imaginary part of third order nonlinear susceptibility of GaAs system is derved with respect to the change in frequency of laser source using classical oscillator model.The result shows that the change in real and imaginary part of third order nonlinear susceptibility increases with decrease in vibrational frequency.

The incident optical intensity has a great influence on real and imaginary part

of the third order nonlinear susceptibility. Consequently it affects the total change in refractive index and absorption coefficient.Now day's optical materials with large third order nonlinear susceptibility are the active areas of research for developing devices for optical limiter and all optical switching devices.

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Chapter 1

Introduction

Considered as fundamental physical quantities that govern nonlinear optical(NLO)phenomena nonlinear susceptibility has been the subject of intense study both theoretically and experimentally since the beginning of nonlinear optics.Nonlinear optics is the study of phenomena that occur as a consequence of the modification of the optical properties of a material system by the presence of light.An optical limiter that is transparent for low input intensities and blocks the output at high intensities is required which is also effective over a wide band of wavelengths[1].

The optical property is based on the imaginary part of the third order nonlinear susceptibility called the nonlinear absorption.On the other hand in transmission based devices materials are required to have large nonlinear refractive index (real part of the third order nonlinear susceptibility)accompanied by minimal absorption losses[2,3].

1.1 Statement of the problem

Recently there is a considerable interest in finding materials having large yet fast nonlinearities. This interest that is driven primarily by the search of materials for

all optical switching and sensor protection application concerns both nonlinear absorption and nonlinear refraction.For optical switching and optical limiting materials are required to have large nonlinear absorption using large power lasers.Nonlinear materials for optical limiting application should have large nonlinearity, high figure of merit,wide dynamical range, and fast response time.

In transmission based devices, materials are required to have large nonlinear refraction accompanied by minimal absorption losses. Due to these important applications it is necessary to study the optical property of materials with respect to different optical parameters. In this thesis the third order nonlinear absorption coefficient and refractive index of GaAs will be solved analytically by using anharmonic oscillator model

1.2 Objectives

1.2.1 General objective

The main aim of this research work is to understand the nonlinear optical properties of GaAs.In doing so,the third order nonlinear absorption coefficient and refractive index will be analyzed numerically using anharmonic oscillator model

1.2.2 Specific objective

To describe the nonlinear optical properties of materials;

To determine the third order nonlinear refractive index of the GaAs ;

To determine the third order nonlinear absorption coefficient of the GaAs.

1.3 Significance of the study

This study helps to understand the magnitude of third order nonlinear absorption coefficient and third order nonlinear refractive index of gallium arsenide (GaAs) at different frequency regime by using anhrmonic oscillator model and there application.

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Chapter 2

Literature Review

2.1 Electromagnetic waves and Maxwell's equations

2.1.1 Electromagnetic waves

Maxwell's equations predict the existence of electromagnetic waves that propagate through space at the speed of light c.Heinrich Hertz confirmed Maxwell's prediction when he generated and detected electromagnetic waves in 1887.That discovery has led to many practical communication systems, including radio, television, radar, and opto-electronics. On a conceptual level, Maxwell unified the subjects of light and electromagnetism by developing the idea that light is a form of electromagnetic radiation. The waves consist of oscillating electric and magnetic fields at right angles to each other and to the direction of wave propagation [2].

The electromagnetic waves are transverse waves.The waves radiated from the oscillating charges can be detected at great distances.Furthermore,electromagnetic waves carry energy and momentum and hence can exert pressure on a surface.The wide range of frequencies covered by electromagnetic waves.For example,radio waves (frequencies of about $10^7 Hz$) are electromagnetic waves produced by oscillating currents

in a radio tower's transmitting antenna. Light waves are a high-frequency form of electromagnetic radiation (about $10^{14}Hz$) produced by oscillating electrons in atoms [3]

2.1.2 Electromagnetic radiation in Vacuum

The electric and magnetic fields are in phase and mutually perpendicular as a consequence of Maxwell's equations.Physically, Electric field results from electric charges or from time dependent magnetic fields, while magnetic fields result from electric currents or time - dependent electric field. Maxwell's equations can be manipulated to give classical wave equations from the time and space dependence of the electric and magnetic field.A significant achievement of this picture was the relation derived between the speed of light c and the product of the electric permittivity ϵ_o and magnetic permeability μ_o of free space [4].

$$c^2\mu_o\epsilon_o = 1 \tag{2.1.1}$$

Maxwells equation are postulates that predict about the physical property of light. In vacuum, they are:

$$\nabla \cdot \mathbf{E} = 0 \tag{2.1.2}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{2.1.3}$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \tag{2.1.4}$$

$$\nabla \times \mathbf{B} = \mu_o\epsilon_o\frac{\partial \mathbf{E}}{\partial t} \tag{2.1.5}$$

where $\varepsilon_0 = 8.854 \times 10^{-12} c^2/N.m^2$ and $\mu_o = 4\pi \times 10^{-7} T.m/A$ After some manipulation Maxwells equation can be recast in the following form to give classical wave equation:

$$\nabla^2 \mathbf{E} = \epsilon_o \mu_o \frac{\partial^2 \mathbf{E}}{\partial t^2}, \tag{2.1.6}$$

$$\nabla^2 \mathbf{B} = \epsilon_o \mu_o \frac{\partial^2 \mathbf{B}}{\partial t^2}. \tag{2.1.7}$$

From the symmetrical relationship between the magnetic and electric field, there are two quantities, the scalar potential ϕ and the vector potential $\vec{A}$,from which $\vec{E}$ and $\vec{B}$ can be derived [5]

$$\mathbf{E} = -\nabla\phi - \frac{\partial \mathbf{A}}{\partial t} \tag{2.1.8}$$

$$\mathbf{B} = \nabla \times \mathbf{A} \tag{2.1.9}$$

Hence the vector potential also obeys the classical wave equation,

$$\nabla^2 \mathbf{A} = \mu_0 \varepsilon_0 \frac{\partial \mathbf{A}}{\partial t^2} \tag{2.1.10}$$

2.1.3 Electromagnetic radiation in material media

Light propagating through a medium or through the vacuum may be described by a transverse wave,where the oscillating electric and magnetic field components are solutions to the Maxwell's equations. Also the nonlinear polarizations, induced in a medium, have to obey the following equations [6]

$$\nabla \cdot \mathbf{D} = \rho \tag{2.1.11}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{2.1.12}$$

$$\nabla \times \mathbf{E} = -(\frac{\partial \mathbf{B}}{\partial t}) \tag{2.1.13}$$

$$\nabla \times \mathbf{H} = \mathbf{j} + (\frac{\partial \mathbf{D}}{\partial t}) \tag{2.1.14}$$

2.2 Introduction of nonlinear optics

Nonlinear optical (NLO) materials play a major role in nonlinear optics and in particular they have a great impact on information technology and industrial applications.Nonlinear optics is the study of phenomena that occur as a consequence of the modification of the optical properties of a material system by the presence of light.Typically,only laser light is sufficiently intense to modify the optical properties of a material system.let us consider how the dipole moment per unit volume or polarization $P(t)$, of a material system depends on the strength $E(t)$ of an applied optical field. 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 [7,2]

$$P(t) = \epsilon_o \chi^{(1)} E(t), \tag{2.2.1}$$

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

$$P(t) = \epsilon_o[\chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + ...] \tag{2.2.2}$$

$$P(t) = P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + ... \tag{2.2.3}$$

The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second-order and third-order nonlinear optical susceptibilities, the nonlinear susceptibilities depend on the frequencies of the applied fields. Second-order nonlinear optical interactions can occur only in noncentrosymmetric crystals-that is, in crystals that do not display inversion symmetry. Since liquids, gases, amorphous solids (such as 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 occur for both centrosymmetric and noncentredsymmetric media.The most usual procedure for describing nonlinear optical phenomena is based on expressing the polarization $P(t)$ in terms of the applied electric field strength $E(t)$,The main reason why the polarization plays a key role in the description of nonlinear optical phenomena is that a time- varying polarization can act as the source of new components of the electromagnetic field [8].

2.2.1 Nonlinear electron oscillator model

Near the beginning of this century H.A Lorentz developed a classical theory of optical properties in which the electrons and ions of matter were treated as simple harmonic oscillators (i.e, springs)subject to the driving force of applied electromagnetic fields, the results obtained therefore formally identical to those of quantum mechanical treatments,although various quantities are interpreting differently in the classical and quantum- mechanical theories.

Lorentz oscillator equation If we assume[9]the nucleus of the atom is much more massive than the electron, we can treat the problem as if the electron-spring system is connected to an infinite mass, which does not move, allowing us to use the mass of the electron, $m = 9 : 11 \times 10^{-31} kg$ depending on the case, this value may be substituted by the reduced or effective electron mass to account for deviations from this assumption.Moreover,the assumption that the binding force behaves like a spring is a justified approximation for any kind of binding, given that the displacement is small enough (meaning that only the constant and linear terms in the Taylor expansion are relevant)[10].

In Lorentz electron oscillator model the equation of motion for an atomic electron in an electric field without considering a damping force is [11];

$$\frac{d^2x}{dt^2} + \omega_0^2 x = \frac{e}{m} E_0 cos\omega t \tag{2.2.4}$$

where,$x(t)$ is the displacement of the electron from its equilibrium position.The elastic restoring force $(-kx)$in equation(2.2.4)is associated with the potential energy varying quadratically with and it is a characteristic of harmonic oscillator.However because of anharmoncity real atomic or molecular oscillators don't follow this exactly or perfectly.The effective potential energy function for the system can be written more realistically as in refereing [12]:

$$V(x) = \frac{1}{2} m\omega^2 x^2 + Ax^3 Bx^4 + Cx^5 + ... \tag{2.2.5}$$

where,V(x)is the potential energy,ω_0 is the reasons frequency,a,b and c is coefficients characteristic of the medium and ω is the filed fragrancy,N is the number of electron. The deriving force in non linear electron oscillator is the negative gradient of equation

(2.2.5) and given by:

$$F(x) = -m\omega_0^2 x - 3Ax^2 - 4Bx^3 - 5Cx^4... \tag{2.2.6}$$

In the limit of elastic restoring force,$a = b = c = ... = 0$ we recover the familiar harmonic oscillator model. The equation of motion in the case of nonlinear oscillator model is:

$$\frac{d^2x}{dt^2} + \omega_o^2 x + \frac{3A}{m}x^2 + \frac{4B}{m}x^3 + ... = \frac{e}{m}E_o \cos\omega t \tag{2.2.7}$$

where,V(x)is the potential energy,m mass of the electron A,B is the constant value of and ω_0 In equation it is reasonable to suppose that the nonlinear terms are much smaller than the linear term. So we seek for perturabative solution of the nonlinear oscillator equation. If we consider the nonlinear electron oscillator equation,

$\frac{d^2x}{dt^2} + \omega_o^2 x + Ax^2 + Bx^3$

$= \frac{e}{m}E_o \cos\omega t x^{(3)}(t)$

$= \frac{-A}{2\omega_o^2}(\frac{e/mE_o}{\omega_o^2-\omega^2})^2 + \frac{1}{\omega_o^2-\omega^2}[e/mE_o - \frac{3B}{4}(\frac{e/mE_o}{\omega_o^2-\omega^2})^3]$

$$\cos\omega + \frac{-a}{2} \times \frac{1}{\omega_o^2 - 4\omega^2}(\frac{e/m}{\omega_o^2-\omega^2})^2 E_o^2 \cos 2\omega t - \frac{B}{4(\omega_o^2 - 9\omega^2)}(\frac{e/mE_o}{\omega_o^2-\omega^2})^3 \cos 3\omega t \tag{2.2.8}$$

An important new effect is evident in this solution. The improved third order nonlinear approximation to the electron displacement $x(t)$ shows that the electron displacement has a term oscillating at fundamental driving field frequency ω,a term oscillating, at second harmonic frequency 2ω , and also a term oscillating at third harmonic frequency 3ω.There is also a static or dc term. It is also possible to write the above equation to a different notation that is convenient for nonlinear optics [13].

$$\mathbf{E}(t) = \mathbf{E}_o \cos\omega t = \Re(\varepsilon_\omega e^{-i\omega t}) = \frac{1}{2}(\varepsilon_\omega e^{-i\omega t} + \varepsilon_\omega^* e^{-i\omega t}) \tag{2.2.9}$$

,employing this,the above solution can be written as;

$$x^{(3)}(t) = x_o \tfrac{1}{2}(x_\omega e^{-i\omega t} + x^*_\omega e^{i\omega t}) + \frac{1}{2}(x_{2\omega} e^{-2i\omega t} + x^*_{2\omega} e^{2i\omega t}) + \frac{1}{2}(x_{3\omega} e^{-3i\omega t} + x^*_{3\omega} e^{3i\omega t}) \tag{2.2.10}$$

$$x_o = \frac{-a}{2\omega_o^2 (\frac{e/m}{\omega_o^2 - \omega^2})^2} |\epsilon_\omega|^2 \tag{2.2.11}$$

$$x_{2\omega} = \frac{-a}{2} \times \frac{1}{\omega_o^2 - 4\omega^2} (\frac{e/m}{\omega_o^2 - \omega^2})^2 \omega_\omega^2 \tag{2.2.12}$$

$$x_\omega = \frac{e/m}{\omega_o^2 - \omega^2} \varepsilon_\omega - \frac{3b}{4(\omega_o^2 - \omega^2)} (\frac{e/m}{\omega_o^2 - \omega^2})^3 \varepsilon_\omega^2 \tag{2.2.13}$$

$$x_{3\omega} = \frac{b}{4(\omega_o^2 - 9\omega_2)} (\frac{e/m}{\omega_o^2 - \omega^2})^3 \varepsilon_\omega^3 \tag{2.2.14}$$

2.2.2 Nonlinear polarization

When one or more electromagnetic waves propagate in a material, the atoms and molecules oscillate not only at the frequencies of the electric field applied, but also at different combinations of those frequencies as a result of the nonlinear response of the medium.The particles of the medium are displaced from their equilibrium positions, so that positive charged particles move in the direction of the field,while the negative charged particles move in the direction opposite to the direction of the applied electric fields. Dipole moments are created because of the displacement between positive and negative charged particles, and the dipole moment per unit volume describes the induced polarization of the medium [14].

In return the medium modifies the optical fields in a nonlinear way. In fact,

all media are nonlinear to a certain degree. The third order nonlinear polarization density P ,the dipole moment per unit volume,is given by:

$$P^{(3)} = -N|e|\chi^{(3)} \tag{2.2.15}$$

If the field at the fundamental frequency ω in the medium is a monochromatic plane wave,

$$E(t) = \frac{1}{2}(\varepsilon_\omega(z)e^{-(\omega t - k_\omega z)} + \varepsilon^*_\omega(z)e^{-i(\omega t - k_\omega Z)} \tag{2.2.16}$$

where, $\varepsilon_\omega = \varepsilon_\omega(z)e^{ik_\omega z}$,$k_\omega = \frac{n(\omega)\omega}{c}$ and the third order polarization density becomes:

$$P^{(3)}(z,t) = P_o^{NL} + \frac{1}{2}[P^L e^{-i(\omega t - k_\omega z)} + P_{2\omega}^{NL} e^{-2i(\omega t - k_\omega z)} + P_{3\omega}^{NL} e^{-3i(\omega t - k_\omega z)}] + c.c \tag{2.2.17}$$

where

$$P_o^{Nl} = \frac{-Nae^3}{2m^2\omega_o^2(\omega_o^2 - \omega^2)}|\varepsilon_\omega(z)|^2 \tag{2.2.18}$$

$$P_\omega^L = \frac{Ne^2/m}{\omega_o^2 - \omega^2}\varepsilon_\omega(z) - \frac{3b}{4(\omega_o^2 - \omega^2)}\frac{Ne^4}{m^3(\omega_o^2 - \omega^2)}\varepsilon_o^3(z) \tag{2.2.19}$$

$$P_{2\omega}^{NL} = \frac{-Nae^3}{2m^2(\omega_o^2 - 4\omega^2)(\omega_0^2 - \omega^2)^2}\varepsilon_\omega^2(z) \tag{2.2.20}$$

$$P_{3\omega}^{NL} = \frac{-Nae^4}{2m^3(\omega_o^2 - 9\omega^2)(\omega_o^2 - \omega^2)^2}\varepsilon_\omega^3(z) \tag{2.2.21}$$

The subscript (L) and (NL) stands for linear and nonlinear. The nonlinear terms vanish when $a = b = ... = 0$ in which case we are return to linear oscillator model. The non linear terms in the polarization arise from the nonlinear properties of the individual atoms or molecules of the medium.

2.3 Nonlinear optical phenomena

Nonlinear optical phenomena can be due to electronic and non electronic process. The former refers to those radiative interactions between the active electron and optical electric field. usually,they are very fast of the order of picoseconds, spatially localized.Examples are second and third order harmonic generation. Non electronic processes are non radiative interactions such as temperature, density, order parameter changes,isomerism and phase transition.[15]

2.3.1 Electronic nonlinear optical process

Let as consider a medium subject to an optical electric field E. The induced polarization can be written as

$$\mathbf{P} = \varepsilon_o(\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{EE} + \chi^{(3)}\mathbf{EEE} + ...) \tag{2.3.1}$$

where, $\chi^{(i)}$ is the electric susceptibility of the order i and ε_o is the permittivity of the free space. Considering centrosymmetric media and keeping up to third order terms in equation (2.3.1) from Maxwell's equation one obtains the refractive index n and the optical absorption α as functions of nonlinearity I of the incident laser beam:

$$n = n_0 + \frac{n_0}{2}|\vec{E}|^2 \tag{2.3.2}$$

$$\alpha = \alpha_o + \beta I \tag{2.3.3}$$

where,n_o and α_o are the linear refractive index and linear optical absorption coefficient respectively,n_2 is named the nonlinear refractive index and β is the nonlinear optical absorption coefficient.The coefficients γ and β are related to the real and imaginary

parts, respectively, of third order electronic susceptibility $\chi^{(3)}$ by [16];

$$\gamma = \frac{1}{n_o^2 \epsilon_o c} \Re(\chi^{(3)}) \tag{2.3.4}$$

$$\beta = \frac{\omega}{n_o^2 \epsilon_o c^2} \Im(\chi^{(3)}) \tag{2.3.5}$$

In electronic nonlinear optical phenomena, the refractive index at a given point on the material depends only on the light intensity at the same point. The electronic nonlinear optical phenomena usually have response time of the order of femto to picoseconds. The macroscopic susceptibility of third order $\chi^{(3)}$ is linearly related to the macroscopic second order hyper polarizablity γ.

2.4 Nonlinear materials

Nonlinear optical (NLO) materials play a major role in nonlinear optics and in particular they have a great impact on information technology and industrial applications. In the last decade,however,this effort has also brought its fruits in applied aspects of nonlinear optics.This can be essentially traced to the improvement of the performances of the nonlinear materials.

The understanding of the nonlinear polarization mechanisms and their relation to the structural characteristics of the materials has been considerably improved. The new development of techniques for the fabrication and growth of artificial materials has dramatically contributed to this evolution. The aim is to develop materials presenting large nonlinearities and satisfying at the same time all the technological requirements for applications such as wide transparency range,fast response and high damage threshold [17].But in addition to the process ability, adaptability and interfacing with other materials improvements in nonlinear effects in devices, led the way

to the study of new nonlinear effects and the introduction of new concepts.Optical solutions, optical switching and memory by nonlinear effects, which depend on light intensity.

Nonlinear materials typically have a distinct crystal structure,which is anisotropic with respect to electromagnetic radiation.The importance of nonlinear optics is to understand the nonlinear behavior in the induced polarization and to analyze and to control its impact on the propagation of light in the matter [18].

2.4.1 Second order nonlinear optical materials and their application

One of the most widely used applications for nonlinear media is the generation of second harmonic radiation at frequency ω is converted into radiation at frequency 2ω within the nonlinear medium. This process generally occurs through virtual transitions where two photons at ω are simultaneously destroyed while a single photon at 2ω is created in a single quantum mechanical process. This process can result in nearly complete conversion of the incident fundamental energy into the second harmonic, and is used extensively in the laboratory to obtain coherent optical radiation with wavelengths other than the fundamental of the fixed-wavelength laser source.

The use of second harmonic generation as a in addition to SHG, second order nonlinearity $\chi^{(2)}$ contributes to many interesting effects like optical rectification, linear electro-optic effect(LEO),parametric oscillation and sum and difference frequency mixing. The second order polarization [19,2] $P^{(2)}$, which is the source of such nonlinearities, can be given as,.

$$P^{(2)}(\omega_3) = \chi^{(2)}(-\omega_3, \omega_1, \omega_2) E_{\omega_1} E_{\omega_2} \tag{2.4.1}$$

where E_{ω_1} is the amplitude of the field at frequency ω_i when $\omega_3 = \omega_2$ and, $\omega_3 = 2\omega$, the polarization, $P(\omega_3 = 2\omega)$, is the source of second harmonic generation (SHG).This effect was first observed by Franken and his co-workers,by focussing the 694.3 nm output from a ruby laser onto a quartz crystal and obtaining a very low intensity output at a wavelength of 347.15 nm. Thus the frequency of the incident light gets doubled which enables one to extend the range of laser wavelengths into the blue and ultraviolet parts of the spectrum.

One important application from SHG is the laser induced nuclear fusion which appears to be more efficient at higher optical frequencies. In recent years, several papers have reported the occurrence of solutions with SHG. When $\omega_3 = 0$, the effect is called optical rectification. These two results are special cases of the process known as sum and difference frequency mixing of the general cases where

$$\omega_3 = \omega_1 + \omega_2 \tag{2.4.2}$$

The left hand side in equation (2.4.2) represents the case of odd-numbered coherence lengths and right hand side in even-numbered ones. If the frequency ω_3 alone is applied, then the two smaller frequencies can build up from noise where ω_1 and ω_2 are known as 'signal' and 'idler' frequencies respectively. A special case that has found many applications is $\omega_1 = 0$ and $\omega_3 = \omega_2 = \omega$, in which case,

$$P^{(2)}(\omega) = \chi^{(2)}(-\omega, 0, \omega) E_o E_\omega, \tag{2.4.3}$$

which is the LEO effect or the Pockel's effect. In this effect,the dc field can be thought of as changing the linear optical susceptibility of the material so that the optical field's propagation parameters are changed by the voltage. Although the input frequency is not modified, the amplitude and phase of the field depend linearly

on the magnitude of the dc field.

It can be seen from equation (2.4.2) that if ω_3 and ω_1 are fixed, then ω_2 is also fixed. However, if only ω_3 is fixed, then the other two are free to range over many values; this effect is known as parametric amplification which is used to produce coherent radiation at frequencies in the ultraviolet region The additional wavelengths made available by harmonic generation and parametric oscillation lead to applications in photochemistry, high resolution spectroscopy, and the remote detection and the identification of atmospheric pollutants by optical radar [20].

2.4.2 Third order nonlinear optical materials and their application

Third-order nonlinear optical effects are the physical basis of a number of applications in future high-capacity communication networks in which ultrafast switching, signal regeneration, and high-speed demultiplexing would be performed all-optically through the use of third order optical nonlinearities.Thus,in recent years,third order nonlinearities have been studied experimentally in a large variety of artificial materials, which include homogeneous bulk glasses and polymers and nano-and mesoscopically structured materials such as semiconductor, quantum-well structures or glasses doped with either semiconductor or metal nanocrystals [21].

Optimization of the design of these materials for a given application depends, to a large extent, on comprehension of the physical mechanisms responsible for the nonlinear response. In this context, it is important to remark that the real and imaginary parts of $\chi^{(3)}$ are the physical parameters that contain the information relative to the microscopic origin of the nonlinearity since they are related to the hyper polarizabilities of the system. However,these quantities are not directly measurable in most cases.

Instead,they are obtained through experimental methods that are sensitive to other macroscopic manifestations of $\chi^{(3)}$, such as phase modulation, nonlinear refraction, wave mixing and nonlinear absorption [22,4].

2.4.3 Nonlinear optical properties in semiconductor materials (GaAs)

In recent years, there is much interest in the characterization of GaAs crystal because of its potential optoelectronics applications in the various fields. For instance, GaAs has caused widespread concern as a semiconductor saturable absorber for its advantages of stable photochemical property, good thermal conductivity, non-degradability and high damage threshold. GaAs-based solar cells also have many advantages, such as high photoelectric conversion efficiency, fine radiation resistance and good performance at high temperatures. As science and technology develops, people always expect to broaden the applications of GaAs.

Doping other elements is one of the most commonly used methods, and is performed by replacing a small amount of anion species in GaAs with isovalent impurities, such as N, P, Sb and Bi. As an important III-V direct-band-gap semiconductor, GaAs has attracted a great deal of interest. It has been characterized with high electron mobility, small dielectric constant, high temperature resistance, and ant radiation and is now widely used in the ultrafast, ultrahigh frequency, low-power devices and circuits. Technically, the electronic structure and optical properties of GaAs can be changed by doping, which provides an effective way to tailor its electronic properties for practical electronic devices. Bismuth in group-V is the heaviest nonradioactive element.

In addition,$GaAs_{1-x}Bi_x$ alloy will decrease the energy gap, extending the application to long-wave emission devices and photovoltaic industry with increasing of the Bi concentrations. The dispersion properties of the second-order nonlinear optical susceptibility of GaAs were first measured by Chang,et al.Their results indicate that the second order susceptibility is correlated to the electronic band structure.A.one photon resonance has been assigned to the direct band gap (E_o).Since then,a few related experiments and theoretical calculations have been conducted by different groups.

The detailed second-order nonlinear optical susceptibilities of GaAs have been calculated from the band structure obtained by empirical methods. On the other hand, studies of symmetry patterns of second-order nonlinear optical processes also reveal the spatial structure of GaAs. With a zinc-blende structure, GaAs carries a single nonzero bulk second-order susceptibility $\chi^{(2)}$, whose contribution to the output radiation is highly anisotropic [23].

2.5 Nonlinear Materials as Optical Limiters and Switching Devices

Optical limiting is a nonlinear optical process in which the transmittance of a material decreases with increased incident light intensity.Optical limiters are one of the most important types of devices used to control the amplitude of high intensity optical pulses. These devices work due to intrinsic properties of the materials used for their fabrication. An ideal optical limiter has a linear transmittance at low input intensities, but above the threshold intensity its transmittance becomes constant. One of the main potential applications of these devices is sensor and eye protection.

All photonic sensors, including the human eye, have a threshold intensity above which they can be damaged,By using the appropriate materials as optical limiters, one can extend the dynamical range of the sensors,allowing them to function optimally at higher input intensities.Now a day there has been a great interest for the development of optical limiting materials to protect optically sensitive devices from laser damage[24,2].

Generally it is expected for the nonlinear materials to have the following characteristics to be used as optical limiter. These are (I) low loss ,(II) high nonlinearity ,(III) fast response time, (IV) large dynamical range and (V) fast spectral response,However, there is no a perfect material that fulfils the mentioned characteristics. Recently one of the active areas of research is towards the improvement of materials for the optical limiters and all optical switching designs. It is possible to improve the optical limiting property of the materials by doping them with suitable guest materials.

An optical limiter that is transparent for low input intensities and blocks the out put at high intensities is required which is also effective over wide band wavelengths.The operation of such a passive optical limiter with a fast response against the nanosecond and ultra fast pulse input is based on the phenomenon of nonlinear absorption parameter is a parameter of importance in determining the optical limiting performance. For photonic switching application a material should have a high value of the effective nonlinear refractive index and the growth and relaxation of the induced refractive index change have to be sufficiently rapid. The material losses that are due to the one photon and multi photon absorption and scattering have to be manageable.[25]

Chapter 3

Materials and Methodology

3.1 Materials

The study is purely theoretical for understanding the nonlinear optical property of materials and for determining the third order nonlinear absorption coefficient and nonlinear refractive index of GaAs.The main sources of literature review are the published articles,books,thesis and dissertations,Matlab soft wares and computers are additional instruments used to accomplice this project.

3.2 Methodology

3.2.1 Analytical

In this thesis one of the method or approach used to solve the problem is analytical method.that is the third order nonlinear absorption coefficient and refractive index of GaAs is obtained using analytical method.

3.2.2 Computational(graphical)

To compare the values of the third order nonlinear absorption coefficient and third order nonlinear refractive index as a function of frequency computational and graphical techniques are employed.Ingeneral the result is interpreted computationally and graphically with the help of MATLAB software.

Chapter 4

Change in third order nonlinear refractive index and absorption coefficient of GaAs using anharmonic oscillator model

The frequency response of the real and imaginary part of third order nonlinearity is studied using anharmonic oscillator model.The change in real and imaginary part of third order nonlinear susceptibility of GaAs system is calculating with respect to the change in frequency of laser source the result shows that the change in real and imaginary part of third order nonlinear susceptibility increases with decrease in vibrational frequency.The incident optical intensity has a great influence on real and imaginary part of the third order nonlinear susceptibility.Consequently it affects the total change in refractive index and absorption coefficient.

Third order nonlinear effects are generally characterized by the nonlinear absorption(β) and nonlinear refractive index n_2 This are described by $n = n_o + \frac{n_2}{2}|\vec{E}|^2$ and $\alpha = \alpha_o + \beta I$where and stands for linear absorption and refractive index respectively.The βandn_2 values are used to evaluate the imaginary ($Im\chi^{(3)}(\omega, \omega, \omega,)$) and real ($Re\chi^{(3)}(\omega, \omega, \omega,)$) parts of third order nonlinearity. In this part of the thesis,the changes in real and imaginary part of third order nonlinear susceptibilities which are induced by incident

optical intensity are studied using classical harmonic oscillator model.

Treatment of third order nonlinear susceptibility using anharmonic oscillator model by using the idea of the Lorentz oscillator we consider a general potential energy V(x) expanded around the equilibrium position of moving charges. The equation of motion is:

$$\ddot{x} + \gamma\dot{x} + \omega_0^2 x + \sum^{\infty_{n=1}} [\frac{\partial^n v(x)}{\partial x^n}] \frac{x^{n-1}}{(n-1)!} = \frac{eE_\omega}{m} \tag{4.0.1}$$

where,$E_o = E_o e^{-i\omega t} + c.c, \gamma$ is the damping constant,e is the electronic charge, m is the mass of electron, ω is the frequency of laser, ω_o is the vibrating frequency of the atom and x is the displacement from equilibrium position. The third order susceptibility $\chi^{(3)}(\omega, \omega, \omega)$ is related to the nonlinear response of the material.The total electric polarization of the medium is,

$$P = Nex \tag{4.0.2}$$

Moreover the total electric polarization in terms of susceptibility can be expressed as:

$$P = \varepsilon_o \chi^{(3)} E \tag{4.0.3}$$

Relating equation (4.0.2) and (4.0.3) the third order susceptibility for third harmonics is,

$$\chi^{(3)}(\omega, \omega, \omega) = \frac{-1}{3!}[\frac{\partial^4 v(x)}{\partial x^4}] \frac{Ne^4/m^3\varepsilon_o}{G(3\omega)G^3(\omega)} + [\frac{\partial^3 v(x)}{\partial x^3}]^2 \frac{Ne^4/2m^3\varepsilon_o}{G(3\omega)G(2\omega)G^3(\omega)} \tag{4.0.4}$$

Where, $G(\omega) = \omega_o^2 - \omega^2 - i\gamma\omega, G(2\omega) = \omega_o^2 - 4\omega^2 - i\gamma\omega, G(3\omega) = \omega_o^2 - 9\omega^2 - i\gamma\omega$, $\frac{\partial^3 v(x)}{\partial x^3}$, and $\frac{1}{3!}[\frac{\partial^4 v(x)}{\partial x^4}]$ are the second and third anharmonic terms and represented by constants

b and a respectively. Simplifying equation (4.0.4), the real part of the third order nonlinear susceptibility is given by:

$$Re[\chi^{(3)}(\omega)] = -\frac{ae^4N}{4\varepsilon_0 m^3}C + \frac{be^4N}{2\varepsilon_0 m^3}(D \times Z - L) \tag{4.0.5}$$

where as,

$$C = \frac{(\omega_0^2-9\omega^2)(\omega_o^2-\omega^2)^3-3\gamma^2\omega^2(\omega_0^2-\omega^2)(\omega_0^2-9\omega^2)-3\gamma^2\omega^2[3(\omega_0^2-\omega^2)^2-\gamma^2\omega^2]}{[(\omega_0^2-\omega^2)^2+\gamma^2\omega^2]^3[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2]}$$

$$D = \frac{1}{[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2][(\omega_o^2-\omega^2)^2+\omega^2\gamma^2]^3[(\omega_0^2-4\omega^2)^2+4\omega^2\omega^2]}$$

$$Z = [(\omega_0^2 - 9\omega^2)(\omega_0^2 - 4\omega^2) - 6\gamma^2\omega^2][(\omega_0^2 - \omega^2)^3 - 3\gamma^2\omega^2(\omega_0^2 - \omega^2)]$$

$$L = [2\gamma\omega(\omega_0^2 - 9\omega^2) + 3\gamma\omega(\omega_0^2 - 4\omega^2)] \times [3\gamma\omega(\omega_0^2 - \omega^2)^2 - \gamma^3\omega^3]$$

and the imaginary part of the third order nonlinear susceptibility is given by;

$$Im[\chi^{(3)}(\omega)] = -\frac{ae^4N}{4\varepsilon_0 m^3}K + \frac{b^2e^4N}{2\varepsilon_0 m^3}(Q \times J + T) \tag{4.0.6}$$

than K,Q,J and T is represent by;

$$K = \frac{3\gamma\omega[(\omega-\omega_0)^3-3\gamma^2\omega^2(\omega_0^2-\omega^2)]+(\omega_0^2-9\omega^2)[3\gamma\omega(\omega_0^2-\omega^2)^2-\gamma^3\omega^3]}{[(\omega_0^2-\omega^2)^2+\omega^2\gamma^2]^3[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2]}$$

$$Q = \frac{1}{[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2][(\omega_0^2-\omega^2)^2+\omega^2\gamma^2]^3[(\omega_0^2-4\omega^2)^2+4\omega^2\gamma^2]}$$

$$J = [3\gamma\omega(\omega_0^2 - \omega^2)^2 - \omega^3\gamma^3][(\omega_0^2 - 9\omega^2)(\omega_0^2 4\omega^2) - 6\gamma^2\omega^2]$$

$$T = [\gamma\omega[2(\omega_0^2 - 9\omega^2) + 3(\omega_0^2 - 4\omega^2)] \times [(\omega_0^2 - \omega^2)^3 - 3\gamma^2\omega^2](\omega_0^2 - \omega^2)]$$

The third order susceptibility $\chi^{(3)}(\omega)$ is related to the change in refractive index

$$\frac{\Delta n^{(3)}(\omega)}{n_r} = Re(\frac{\chi^{(3)}(\omega)}{2n_r^2}) \tag{4.0.7}$$

where,n_r is the index of refraction and$\chi^{(3)}(\omega)$is the third order susceptibility.With respect to equation (4.0.5) and (4.0.7) we obtain the change in third order nonlinear refractive index as

$$\frac{\Delta n^{(3)}(\omega)}{n_r} = -\frac{ae^4N}{8n_r^2m^3}U + \frac{b^2e^4N}{4\varepsilon_0 n_r^2 m^3}(H \times W - V) \tag{4.0.8}$$

than U,H,W and V is represent by equation of

$$U = [\frac{(\omega_0^2-9\omega^2)(\omega_0^2-\omega^2)^3-3\gamma^2\omega^2(\omega_0^2-\omega^2)(\omega_0^2-9\omega^2)-3\gamma^2\omega^2[3(\omega_0^2-\omega^2)^2-\gamma^2\omega^2]}{[(\omega_0^2-\omega^2)^2+\gamma^2\omega^2]^3[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2]}$$

$$H = \frac{1}{[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2][(\omega_0^2-\omega^2)^2+\omega^2\gamma^2]^3[(\omega_0^2-4\omega^2)^2+4\omega^2\gamma^2]}$$

$$W = [(\omega_0^2 - 9\omega^2)(\omega_0^2 - 4\omega^2) - 6\gamma^2\omega^2][(\omega_0^2 - \omega^2)^3 - 3\gamma^2\omega^2(\omega_0^2 - \omega^2)]$$

$$V = [2\gamma\omega(\omega_0^2 - 9\omega^2) + 3\gamma\omega(\omega_0^2 - 4\omega^2)] \times [3\gamma\omega(\omega_0^2 - \omega^2)^2 - \gamma^3\omega^3]$$

The change in third order nonlinear refractive index is analytically and numerically determined.The variation of the change in third order nonlinear refractive index versus pump frequency with respect to vibrational frequency ($\omega_o = 1.015 \times 10^{15} rad/s, \omega_o = 1.02 \times 10^{17} rad/s, \omega_o = 1.025 \times 10^{15} rad/s and \omega_o = 1.03 \times 10^{15} rad/s$) are Moreover the imaginary part of third order nonlinear susceptibility or the change in third order nonlinear absorption coefficient is described by using anharmonic oscillator model:

$$\alpha^{(3)}(\omega) = -\frac{2\pi\omega ae^4N}{n\varepsilon_0^2 c4m^3}P + \frac{2\pi\omega b^2e^4N}{n\varepsilon_0^2 c2m^3}(F \times \rho) \tag{4.0.9}$$

Where,P,F and ρ are

$$\delta = \frac{3\gamma\omega[(\omega-\omega_0)^3-3\gamma^2\omega^2(\omega_0^2-\omega^2)]+(\omega_0^2-9\omega^2)[3\gamma\omega(\omega_0^2-\omega^2)^2-\gamma^3\omega^3]}{[(\omega_0^2-\omega^2)^2+\omega^2\gamma^2]^3[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2]}$$

$$F = \frac{1}{[(\omega_0^2-9\omega^2)^2+9\omega^2\gamma^2][(\omega_0^2-\omega^2)^2+\omega^2\gamma^2]^3[(\omega_0^2-4\omega^2)^24\omega^2\gamma^2]}$$

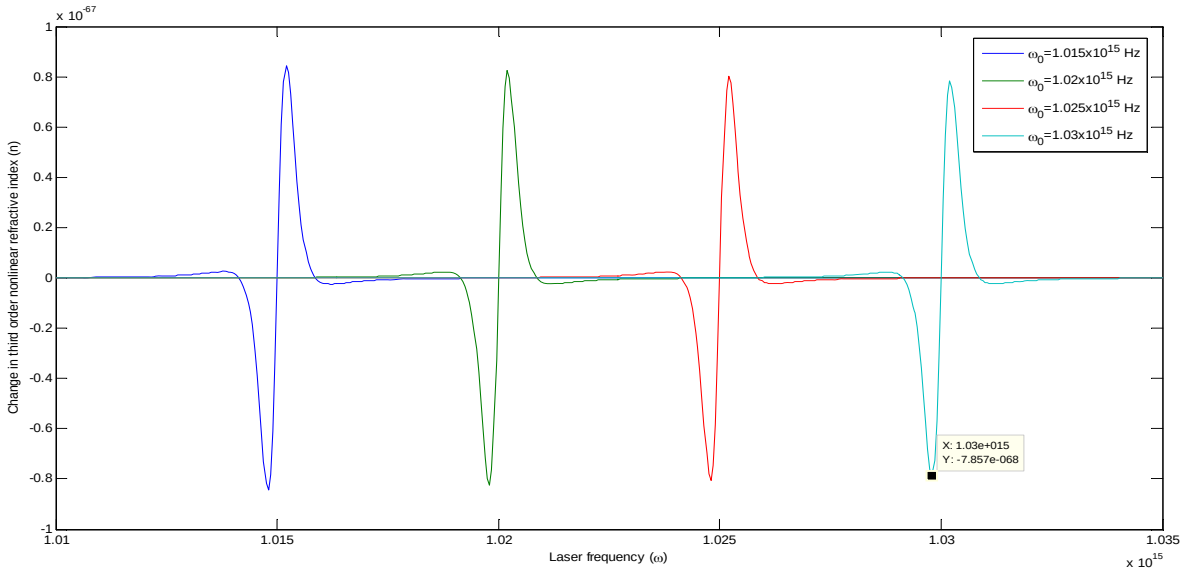

Figure 4.1: *Variation of the change in third order nonlinear refractive index versus laser frequency with* ($\omega_0 = 1.015 \times 10^{15} rad/s, \omega_0 = 1.02 \times 10^{15} rad/s, \omega_0 = 1.025 \times 10^{15} rad/s and \omega_0 = 1.02 \times 10^{15} rad/s$)

$$\rho = ([3\gamma\omega(\omega_0^2 - \omega^2)^2 - \omega^3\gamma^3][(\omega_0^2 - 9\omega^2)(\omega_0^2 - 4\omega^2) - 6\gamma^2\omega^2] + \gamma\omega[2(\omega_0^2 - 9\omega^2) + 3(\omega_0^2 - 4\omega^2)] \times [(\omega_0^2 - \omega^2)^3 - 3\gamma^2\omega^2](\omega_0^2 - \omega^2))$$

The imaginary part of the change in third order nonlinear susceptibility or the change in third order nonlinear absorption coefficient is observed analytically as well as numerically. The variation of the change in third order nonlinear absorption coefficient versus pump frequency with vibrational frequency ($\omega_0 = 1.015 \times 10^{15} rad/s, \omega_0 = 1.02 \times 10^{15} rad/s, \omega_0 = 1.025 \times 10^{15} rad/s and \omega_0 = 1.03 \times 10^{15} rad/s$) The variation of the change in third order nonlinear refractive index versus the frequency of laser source is shown in Fig.1. From the figure one can observe that the change in third order nonlinear refractive index exhibits blue shift for the increment of vibrational

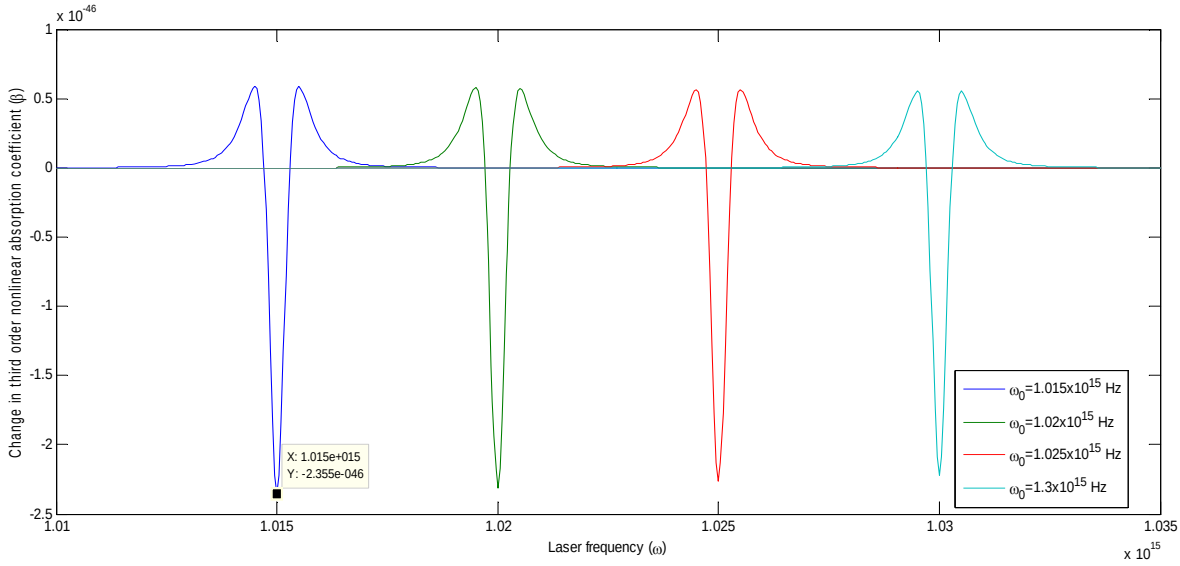

Figure 4.2: *Variation of the change in third order nonlinear absorption coefficient versus laser frequency with* ($\omega_0 = 1.015 \times 10^{15} rad/s, \omega_0 = 1.02 \times 10^{15} rad/s, \omega_0 = 1.025 \times 10^{15} rad/s$ *and* $\omega_0 = 1.03 \times 10^{15} rad/s$)

resonance frequency.

However; its threshold magnitude decreases with increment of resonance frequency.Moreover the variation of the change in third order nonlinear absorption coefficient against the frequency of laser source is described in Fig.2.This figure clearly reveals that the change in third order nonlinear absorption coefficient has a maximum value at resonance frequency and then decreases and vanishes far away from the resonance frequency.Hence, power loss due to the imaginary part of the third order nonlinear susceptibility can be maximized by operating the pump at a frequency near to the resonance frequency. This behavior may be very important while designing optical limiters.

Furthermore Fig.1 and Fig.2 illustrate,that as a vibrational frequency decreases, both

the maximum change in the real and imaginary part of third order nonlinear susceptibility will move to the left of the curve. This predicts reducing the vibrational frequency induces red shift of the resonant absorption.Another observed feature is the threshold magnitude of the third order nonlinear absorption coefficient decreases as a vibrational frequency of the system increases.

Chapter 5

Conclusion

The change in third order nonlinear refractive index and absorption coefficient are studied employing nonlinear anharmonic oscillator model. The result clearly shows that this model gives approximately good signature for describing the change in third oreide nonlinear refractive index and absorption coefficient. The obtained value is negative and it is the cause for the decrease in total change in real and imaginary part of the susceptibility. The result shows that as a vibrational resonant frequency increases the threshold value of the change in third order absorption coefficient and refractive index exhibits blue shift. However the magnitude of the threshold value of the third order nonlinear refractive index and absorption coefficient decreases with increment of vibrational frequency.

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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.

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