Second-order optical susceptibility in doped III-V piezoelectric semiconductors in presence of magnetostatic field

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A detailed analytical investigation of second-order optical susceptibility has been made in moderately doped III–V weakly piezoelectric semiconductor crystal, viz. n-InSb, in the absence and presence of an external magnetostatic field, using the coupled mode theory. The second-order optical susceptibility arises from the nonlinear interaction of pump beam with internally generated density and acoustic perturbations. Effect of doping concentration, magnetostatic field and pump intensity on second-order optical susceptibility of III-V semiconductors has been studied in detail. The numerical estimates are made for n-type InSb crystals duly shined by pulsed 10.6 μ m CO₂ laser and efforts are made towards optimizing the doping level, applied magnetostatic field and pump intensity to achieve large value of second-order optical susceptibility and alteration of its sign. The enhancement and change of sign of second-order optical susceptibility in weakly piezoelectric III-V semiconductor under proper selection of doping concentration and externally applied magnetostatic field confirms them as potential candidate materials for the fabrication of nonlinear optical devices. In particular, at B₀=14.1 T, the second-order susceptibility was found to be 3.4×10^{-7} (SI unit) near resonance condition.

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1. Introduction

The study of nonlinear optical susceptibility (NLOS) provides important information about optical properties of the materials and also plays pivotal role in fabrication of efficient optoelectronic devices, to name a few, amplifier, oscillator, frequency converter, sensors [1,2], pulse squeezing and signal processing, etc. [3]. In addition, change of sign of NLOS leads to phenomena like self focusing/defocusing, amplification/attenuation and are being utilized in fabrication of optical switches, optical limiters and amplifiers [4, 5].

The choice of a nonlinear medium and operating wavelength are crucial aspects in design and fabrication of efficient opto-electronic devices. Amongst different types of nonlinear medium, semiconductors offer considerable flexibility for fabrication of optoelectronic devices because: (i) the large number of free electrons/holes available as majority charge carriers in doped semiconductors manifest many more exciting nonlinear optical processes [6]; (ii) carrier relaxation times can be altered through design of materials and device structures; (iii) large optical nonlinearities in the vicinity of band gap resonant transitions [7, 8]; (iv) either absorption changes or refractive index changes can be utilized; (v) devices may operate either at normal incidence or in waveguides; (vi) devices are integrable with other optoelectronic components.

So far, resonant optical nonlinearities, due to their large magnitudes, have been implemented to improve the efficiency of nonlinear optical devices [9]. However,

speed of devices, based on resonant nonlinearities are slow. Because resonant optical nonlinearities depend upon population changes during the real transition and energy built up as well as relaxation times of charge carriers. In contrast, the semiconductors nonresonant optical nonlinearities (SNON), which occur in the off-resonant transition regime, are typically of much smaller in magnitude but exhibit much faster response times, since they do not involve in the generation and relaxation of charge carriers.

Ultrafast optical switching devices play an increasingly important role in modern optical communication systems where signals are transmitted at high speed and high-bit-rate. Although many materials with higher nonlinear refractive index have been reported [10], but there is a need to explore alternatives, particularly those compatible with semiconductor technology, due to the rapid growth of planar devices such as wavelength division multiplexers and add-drop filters in advanced networks. To achieve compact low-power highspeed switching devices, a large optical nonlinearity with fast response time is required. In addition, materials with minimal loss and absorption at the operating wavelength, a fast response, low toxicity with highly thermal and mechanical stability are preferred.

Above discussion clearly manifests that SNON is potential phenomena in high speed optical communication systems and its enhancement and manipulation are important issues to improve the efficiency and performance of devices based on it. Doping (free carriers), composition and micro-structuring have been mostly exploited for enhancing SNON and the performance of related optoelectronic devices [11, 12]. In addition, optical nonlinearities of semiconductors can be easily modified by externally applied electric and magnetic fields. This mechanism is being exploited to understand the mechanisms involved in several nonlinear processes such as electro-optic and magneto-optic effects [9]. Several research groups [13, 14, 15] have observed a large enhancement in surface field terahertz (THz) emission from a variety of III–V semiconductors when subjected to an externally applied magnetic field. Apart from this, noncentrosymetric semiconductors support many new modes and offer many new channels for scattering in the presence of external fields, which have been utilized in optoacoustic diagnostics [16].

It is well known fact that noncentrosymetric crystals possess both even ($\chi^{(2)}, \chi^{(4)}, -$) and odd ($\chi^{(3)}, \chi^{(5)}, -$ -) order optical nonlinearities. The second-order optical susceptibility ($\chi^{(2)}$) is the lowest order optical nonlinearity and is larger than the other higher-order optical susceptibilities. The origin of fundamental nonlinear phenomena, (to name a few; frequency conversion, frequency tuning, parametric oscillation/ amplification, filtration, pulse compression and cascaded nonlinear process), lies in $\chi^{(2)}$ and hence $\chi^{(2)}$ has been utilized in the fabrication of fundamental optoelectronic devices such as frequency converter, parametric oscillator/ amplifier and switch ete [17].

Recently, a long period linear grating are imprinted on semiconductor waveguide and frequency conversion is experimentally demonstrated [18]. Using the finitedifference beam propagation method, nonlinear propagation and wave mixing characteristics of pulses in semiconductor optical amplifiers are also analyzed [19]. Koshevaya et. al. [20] discussed two different mechanisms of amplification of hypersound waves in bulk n-GaAs, namely (i) travelling wave mechanism (piezoelectric effect, deformation potential and electrostriction), (ii) negative differential mobility (Gunn effect) and proposed realistic construction of filters and delay lines etc. for communication and control system. Optical second harmonic generation has been induced in single and polycrystalline InAs by acoustic wave and it was found that $\chi^{(2)}$ depends on acoustic power [21]. A layered periodic structure of III-V semiconductor-plasma has been utilized for significant increase in efficiency of nonlinear interaction near nonlinear resonances [22].

To the authors knowledge, it appears from available literature that no systematic attempt has so far been made to explore the influence of free carrier concentration, externally applied magnetostatic field and excitation intensity on second-order susceptibility in weakly piezoelectric n-type III–V semiconductors and which is the subject of the present article. Using the coupled-mode theory, expression for effective second-order optical susceptibility in the III-V semiconducting crystals is derived. Finally, exhaustive numerical analysis is performed with a set of data appropriate for weakly piezoelectric semiconductor- plasma (n-InSb) duly irradiated by a 10.6 μ m CO₂ laser to establish the validity

of the present model. The results indicate that the secondorder optical susceptibility can be tailored by varying carrier concentration, externally applied magnetic field and pump intensity; and open up possibilities of fast nonlinear optical devices.

2. Theoretical formulation

The present section deals with the determination of complex effective optical susceptibility in weakly piezoelectric doped III-V semiconductors plasma under off-resonant transition regime in the absence and the presence of external applied magnetostatic field. Here, it is worth pointing out that there are many advantages of choosing weakly piezoelectric homogeneous III-V semiconductor possessing isotropic nondegenrate parabolic band structure, as opposed to strong piezoelectric semiconducting crystals such as CdS and ZnO. In particular, in narrow band-gap weakly piezoelectric semiconductors such as InSb and InAs, the high mobility of electrons (due to their low effective mass) allows one to work at drift velocities several times the sound velocity and thus keep the effects of inhomogeneities to a minimum [23].

The model used in the analysis is the well-known hydrodynamic model of the homogeneous one component (viz., n-type) semiconducting-plasma, satisfying the condition $k_a \sim 1$ (k_a and l being the acoustic wave number and the electron mean free path, respectively). This condition implies that the sound wavelength is much greater than the average distance the electron travels between collisions so that the motion of the carriers under the influence of the external field is averaged out. In addition, it allows neglecting the nonuniformity of the high frequency electric field under the dipole approximation [24]. We also consider relaxation-time and large quantum number approximation in which the periodic motion in the plane normal to the axis of magnetostatic field may be neglected and the effect of magnetic field on the electronic properties can be obtained readily from a simple generalization of the classical model [25].

Let us assume the pump (laser) energy $(\hbar \omega_n)$ is smaller than the band gap energy of semiconductor and electric associated field is defined as $E_p(x,t) = E_p \exp[i(k_p x - \omega_p t)]$. Further we assume the pump to be an infrared pulsed laser with the pulse duration much larger than the acoustic damping time such that the interaction is treated to be of a steady-state type. The coupled mode approach is being used to determine effective nonlinear optical susceptibilities of semiconducting plasma. Here it is worth pointing out that in the treatment of coupled wave problems, the classical description is even more appropriate because the decay or amplification of the waves depends on the relative phases among them. Besides, the classical electromagnetic approach yields valuable information regarding the threshold value of pump electric field and gain mechanism of stimulated scattering phenomena like stimulated Brillouin and Raman scattering [26, 27].

For electromagnetic treatment, we assume there are many photons in the pump wave and it can be described by plane wave. The pump wave produces stress in the medium and the linear relationship between stress and electric field is described by piezoelectricity. The origin of

piezoelectricity lies in the first order force $(f_1 = \beta \frac{\partial E}{\partial x})$,

where β is piezoelectric coefficient). Here it is worth mentioning that the piezoelectric behavior of the ferroelectric materials has been treated extensively due to their applications in sensors, electromechanical actuator and acoustic transducers [28].

The schematic scheme of the mixing of pump wave, acoustic wave and scattered wave in the presence of an external magnetostatic field is shown in Fig. 1 under one dimensional configuration. The magnetostatic field (B_0) is applied along the z direction. We assume the pump wave propagates along x-direction and incident on the crystal. Let us assume the electric field of pump wave produces longitudinal acoustic wave $u(x, t) = u_0 \exp[i(k_a x - \omega_a t)]$ and in turn it scatters the pump wave. We further assume that stoke component of scattered wave, say $E_s(x, t) = E_s \exp[i(k_s x - \omega_s t)]$, also propagates in the opposite direction (backward scattering) of the pump wave. In general, noncentrosymetric crystals under an externally applied electric and magnetic field, the piezoelectricity coefficients are no longer isotropic and therefore offdiagonal components of the susceptibility tensor are nonzero. The linear and nonlinear response of such systems should be treated in three-dimensional space and will be the subject of future publication.



Fig. 1. Schematic diagram for interaction of pump with III-V semiconducting crystal in the presence of magnetostatic field.

In the present study, for simplicity, we consider generation of longitudinal acoustic wave in a cubic media possessing $\overline{4}$ 3m symmetry and for such mode the piezoelectric and photo-acoustic tensor may be reduced to single component [23, 29]. It is well known fact that the piezoelectric force drive the acoustic wave in the crystalline medium [30]. The presence of acoustic wave (\vec{k}_a, ω_a) modulates the optical dielectric constant and thus can cause an exchange of energy between the electromagnetic waves whose frequencies differ by an amount equal to acoustical frequency. The momentum and energy transfer between electromagnetic and acoustic waves can be described by phase matching conditions:

$$\hbar k_p = \hbar k_s + \hbar k_a$$
 and $\hbar \omega_p = \hbar \omega_s + \hbar \omega_a$.

Let us start with equation of lattice vibration. If the deviation be u(x, t), then the strain can be expressed as $\partial u / \partial x$. After including the effects of piezoelectricity, the modified equation of motion for u(x, t) of lattice vibrations in the presence of a magnetostatic field may be given as

$$\frac{\partial^2 u}{\partial t^2} - \frac{C}{\rho} \frac{\partial^2 u}{\partial x^2} + 2\Gamma_a \frac{\partial u}{\partial t} = \frac{\beta}{\rho} \frac{\partial E}{\partial x}$$
(1)

Let v_o is the oscillatory fluid velocity of an electron of scalar effective mass m and charge -e. The oscillatory electrons, due to the oscillating pump electric field, feed energy to the lattice vibration of the medium and when electrons are further accelerated by an external magnetic field such that $v_0 >> v_a$ (near the resonance between the stokes wave and the modified cyclotron frequency) which results in strong interaction between electron fluid velocity and lattice vibration. Here v_a is the velocity of the acoustic wave. It is a well-known fact that the effective mass of an electron in III-V semiconductors is smaller than the effective mass of a hole and therefore the drift velocity of a hole is less than the drift velocity of an electron and hence the contribution of the drift velocity of a hole to free carrier nonlinearity can be neglected. ρ , C, Γ_a and Ebeing the mass density, material's elastic constant, acoustic phonon damping parameter and space charge electric field of the medium, respectively.

The other basic equations of present formulation are:

$$\frac{\partial \vec{v}_o}{\partial t} + v \vec{v}_o = -(e/m)(\vec{E}_p + \vec{v}_o \times \vec{B}_o)$$
(2)

$$\frac{\partial \vec{v}_1}{\partial t} + v\vec{v}_1 + \vec{v}_0 \times (\nabla . \vec{v}_1) = -(e/m)(\vec{E} + \vec{v}_1 \times \vec{B}_0) \quad (3)$$

$$\frac{\partial n_1}{\partial t} + n_0 \frac{\partial v_1}{\partial x} + n_1 \frac{\partial v_0}{\partial x} + v_0 \frac{\partial n_1}{\partial x} = 0$$
(4)

$$P = \pm \beta \left(\frac{\partial u}{\partial x} \right) \tag{5}$$

$$\frac{\partial E}{\partial x} = -\frac{n_{\rm l}e}{\varepsilon} - \frac{1}{C}\frac{\partial P}{\partial x} \tag{6}$$

Eqs. (2) and (3) are the zeroth- and first-order electron moment transfer equations under the influence of magnetic field. v_0 and v_1 are, respectively, the zeroth- and firstorder oscillatory fluid velocities of the electron, ν being the electron collision frequency. Eq. (4) is the continuity equation, where n_0 and n_1 are the initial and perturbed electron concentrations in the n-type-doped semiconductor, respectively. Eq. (5) describes induced polarization due to piezoelectricity in the presence of an external magnetic field. In Poisson's eq. (6), ε is the dielectric constant of the crystal material.

Now we shall concentrate on obtaining an expression for perturbed carrier-density due to piezoelectric force. Differentiating Eq. (4) and then substituting the first-order differential coefficient of the equilibrium and perturbed electron fluid velocities through Eqs. (2) and (3) and induced electric field through Eq. (4).

Under rotating-wave approximation, using eqs. (1-6), expression for the density perturbation is obtained as

$$\frac{\partial^2 n_1}{\partial t^2} + v_{eff} \frac{\partial n_1}{\partial t} + \frac{n_1 e A_0}{m} \frac{\partial E_{0x}}{\partial x} \mp \frac{e A_0 E_{0x}}{m} \frac{\partial n_1}{\partial x} \mp \frac{n_0 e A_1}{m} \frac{\partial E_x}{\partial x} - n_0 \frac{\partial v_0}{\partial x} \frac{\partial v_1}{\partial x} = 0$$
(7)

In deriving eq. (7), higher order terms like $v_0 \frac{\partial^2 n_1}{\partial x \partial t}$ and

 $n_0 v_0 \frac{\partial^2 v_{1x}}{\partial x}$ has been neglected.

Using above formulations and after mathematical simplifications, we obtain

$$\frac{\partial^2 n_1}{\partial t^2} + v_{eff} \frac{\partial n_1}{\partial t} + \Omega_p^2 n_1 A_1 \mp \frac{i\beta^2 k_a^3 \Omega_p^2 A_i E_a}{\rho e \Delta_a^2} \pm \frac{ik_p \Omega_p^2 E_p n_i e_1}{\Delta_{p1}^2} \mp \frac{\beta^2 k_p \Omega_p^2 E_p k_a^3 E_a}{\rho m \Delta_a^2 \Delta_{p1}^2} - e_1 A_o E_p \frac{dn_1}{dx} - e_1 in_1 A_o k_p E_p = 0$$
(8)

where $\Omega_p = (n_0 e^2 / m\varepsilon)^{1/2}$ (electron-plasma frequency), $\omega_c = (e/m)B_0$ (electron-cyclotron frequency), $e_1 = e/m$, $\delta_p = v - i\omega_p$, $\delta_1 = v - i\omega_s + ik_s v_0$, $A_0 = 1 - (\omega_c^2 / (\delta_p^2 + \omega_c^2)))$, $A_1 = 1 - (\omega_c^2 / (\delta_1^2 + \omega_c^2))$, $v_{eff} = v + ik_p v_o$, $\Delta_a^2 = -\omega_a^2 + v_a^2 k_a^2 - 2i\Gamma_a \omega_a$ and $\Delta_{p1}^2 = \frac{(\delta_p^2 + \omega_c^2)(\delta_1^2 + \omega_c^2)}{\delta_p \delta_1}$

The density perturbation n_1 oscillate at induced wave frequency components (i.e. ω_a and ω_s) and that can be expressed as: $n_1 = n_{1s}(\omega_a) + n_{1f}(\omega_s)$, where n_{1s} (lowfrequency component) is associated with the acousticwave vibration at ω_a and the n_{1f} (high-frequency component) oscillates at electromagnetic wave frequencies $(\omega_p \pm \omega_a)$. The higher-order terms with frequencies $\omega_{s,q} (= \omega_p \pm q \omega_a)$, for $q \ge 2$ will be offresonant and are neglected in the study of second-order nonlinearities in doped semiconductors. Now we are interested to obtain first-order backward Stokes component of nonlinear current density $(J_{NL}(\omega_s))$ of the piezoelectric phase matching conditions, low and high frequency density perturbation components can be obtained from eq. (8) as

$$\frac{\partial^2 n_{1s}}{\partial t^2} + v_{eff} \frac{\partial n_{1s}}{\partial t} + \Omega_p^2 n_{1s} A_1 \mp \frac{i\beta^2 \Omega_p^2 A_1 k_a^3 E_a}{\rho e \Delta_a^2} - ik_a (e/m) A_0 n_{1f}^* E_p = 0$$
(9a)

and

$$\frac{\partial^2 n_{1f}}{\partial t^2} + v_{eff} \frac{\partial n_{1f}}{\partial t} + \Omega_p^2 n_{1f} A_1 + i e_1 k_p \Omega_p^2 \Delta_{p1}^2 E_p n_{1s}^* \mp \frac{\beta^2 k_p \Omega_p^2 k_a^3 E_p E_s^*}{\rho m \Delta_a^2 \Delta_{p1}^2} - i e_1 A_0 k_s n_{1s}^* E_p = 0$$
(9b)

Eq. (9) reveal that the density perturbation components are coupled to each other via the pump electric field. By solving simultaneous eqs. 9(a) and 9(b), expression of n_{1s} and n_{1f} can be obtained as well as their values may be computed by knowledge of material parameters and electric field amplitudes. The expression of n_{1s} is obtained as

$$n_{1s}^{*} = -\frac{i\beta^{2}\Omega_{p}^{2}k_{a}^{3}E_{a}^{*}}{\rho\Omega_{pl}^{2}} \left[\pm \frac{A_{1}^{*}}{e\Delta_{a}^{2*}} \mp \frac{eA_{p}^{*}k_{p}k_{a}\left|E_{p}\right|^{2}}{m^{2}\Delta_{a}^{2}\Omega_{pd}^{2}\Delta_{p1}^{2}} \right]$$
(10)
$$\Omega_{pl}^{2} = \Omega_{p}^{2}A_{1} - \omega_{a}^{2} - iv_{eff}\omega_{a}, \text{ and}$$
$$\Omega_{pd}^{2} = \Omega_{p}^{2}A_{1} - \omega_{s}^{2} - iv_{eff}\omega_{s}.$$

From eq. (10) magnitude of density perturbation (n_{1s}) can be computed in weakly doped III-V semiconductors both in the absence and presence of externally applied transverse magnetostatic field.

We now address ourselves to the theoretical formulations for the nonlinear polarization at Stoke's frequency arises due to the nonlinear induced current density. The backward Stoke's component of the nonlinear current density can be given as

$$J_{NL}(\omega_s) = n_{1s}^* e v_0 + n_0 e v_1.$$
(11a)

Treating the induced nonlinear polarization as a time integral of the nonlinear current density, one may express

$$P_{eff}(\omega_s) = \int J_{NL}(\omega_s) dt .$$
 (11b)

After further solving effective second-order polarization is obtained as

$$P_{eff} = \left[\pm \frac{i\beta^2 \Omega_p^2 k_a^3 A_1^* e_l \delta_p E_a^* E_p}{\rho \Delta_a^{z^*} \Omega_{pl}^2 (\delta_p^2 + \omega_c^2)} \mp \frac{i\beta^2 \Omega_p^2 k_a^3 e_l^3 A_p^* k_p k_a \delta_p \left| E_p \right|^2 E_a^* E_p}{\rho \Delta_a^2 \Omega_{pd}^2 \Omega_{pd}^2 \Omega_{pl}^2 (\delta_p^2 + \omega_c^2)} \right]$$
(12)

Finally effective second-order optical susceptibility may be obtained as

$$\chi_{eff}^{(2)}(\omega_s) = \pm (\chi^{(2)}(\omega_s))_{\beta} \mp (\chi^{(2)}(\omega_s))_{\beta,I}$$
(13)

where

$$(\chi^{(2)}(\omega_s))_{\beta} = \frac{i\beta^2 \Omega_p^2 k_a^3 A_1^* e_1 \delta_p}{\varepsilon_0 \rho \Delta_a^{2^*} \Omega_{pl}^2 (\delta_p^2 + \omega_c^2)}$$

$$(\chi^{(2)}(\omega_s))_{\beta,I} = \frac{i2\beta^2\Omega_p^2 k_a^3 e_1^3 A_p^* k_p k_a \delta_p I_p}{\varepsilon_0^2 \eta c \rho \Delta_a^2 \Omega_{pd}^2 \Omega_{pl}^2 \Delta_{p1}^2 (\delta_p^2 + \omega_c^2)}$$

with
$$I_p = \frac{1}{2} \eta \varepsilon_0 c \left| E_p \right|^2.$$

In the forthcoming section eq. (13) is being utilized for detailed analysis.

3. Results and deiscussion

The analytical result obtained has been applied to moderately n-type doped III-V semiconductor such as n-InSb. To make the estimation compatible with requirement such as off-resonant laser excitation, we consider the irradiation of n-InSb sample by 10.6 μ m CO₂ at 77 K. It is worth pointing that at liquid nitrogen temperature (77 K), the dominant mechanism for transfer of momentum and energy of the electron in scattered mode is due to the acoustic phonon scattering in semiconductors [31, 32]. The following material parameters are taken as representative values to establish the theoretical formulation:

$$\begin{split} \omega_p &= 1.78 \times 10^{14} \ \text{s}^{-1}, \ \omega_a = 2 \times 10^{11} \ \text{s}^{-1}, \ \omega_s = 1.77 \times 10^{14} \\ \text{s}^{-1}, \ m &= 0.014 m_0 \ (m_0 \ \text{being} \ \text{the rest} \ \text{mass} \ \text{of} \ \text{an} \\ \text{electron}), \ \epsilon_l &= 15.8, \ \rho = 5.8 \times 10^3 \ \text{kgm}^{-3}, \ \beta = 0.054 \ \text{Cm}^{-2}, \\ \mathcal{V} &= 3 \times 10^{11} \ \text{s}^{-1}, \ v_a = 4 \times 10^3 \ \text{ms}^{-1}, \ k_a = 5 \ x \ 10^7 \ \text{m}^{-1}, \ \Gamma_a = 2 \\ x \ 10^{10} \ \text{s}^{-1} \end{split}$$

Eq. (13) shows that effective second-order nonlinear optical susceptibility $(\chi^{(2)}(\omega_s))_{eff})$ at Stokes frequency is a complex quantity. The present theoretical model explains the dependence of $(\chi^{(2)}(\omega_s))_{eff}$ on controllable physical parameters such as wave number/frequency, free carrier concentration (through plasma frequency, Ω_p), external applied magnetostatic field (through cyclotron frequency, ω_c) and pump intensity (I_p). Eq. (13) is used to study $(\chi^{(2)}(\omega_s))_{eff}$ by varying controllable parameters: n₀, B₀ and I_p. Efforts are made to find out appropriate values of controllable parameters and to explore the possibility of fast optoelectronic devices based on secondorder nonlinearity. Eq. (13) reveals that $(\chi^{(2)}(\omega_s))_{eff}$ is finite if β (piezoelectric coefficient) is finite and is the precondition for the occurrence of second-order nonlinearity. The first term of this equation arises solely due to piezoelectricity (first-order force) while the second term is finite if both piezoelectricity and pump intensity are finite. The second term may be defined as intensity-dependentsecond-order susceptibility.

Using straight forward ion for real and imaginary parts of $(\chi^{(2)}(\omega_s))_{eff}$ can be obtained. It is well known fact that real and imaginary part of optical susceptibility describes refraction and gain/absorption phenomena in the crystal, respectively. By knowledge of propagation of light in refractive medium various optical devices have been designed and fabricated such as optical wave guide, filters, amplifiers, coupler etc [13].

Figs. 2 and 3 depict the behavior of real part of eq. (13). Fig. 2 shows behavior of the second-order susceptibility $((\chi_r^{(2)}(\omega_s))_{\beta})$, obtained from the first term. In this figure externally applied magnetostatic field (B_0) and free carrier concentration (n_0) varied simultaneously. The figure clearly shows substantial enhancement of $(\chi_{\rm r}^{(2)}(\omega_{\rm s}))_{\beta}$ as well as change in its sign. This typical behavior arises due to two resonance conditions: (i) $\omega_{\rm s}^2 = (\omega_{\rm c}^2 + {\rm v}^2)/(1 - (\Omega_{\rm p}^2/4\omega_{\rm a}^2))$ (resonance between scattered stokes wave frequency and modified electroncyclotron wave frequency) and (ii) $\omega_s^2 = \omega_c^2$ (resonance between scattered stokes wave frequency and electroncyclotron frequency). The condition at which $(\chi_r^{(2)}(\omega_s))_{\beta}$ changes sign is referred to as 'dielectric anomaly' or 'cut-off' and the excitation near resonance condition is called anomalous dispersion [33]. Near the resonance conditions, magnetostatic field dependent drift velocity becomes many times larger than the acoustic wave velocity and due to which more energy is transferred from the carrier wave to the acoustic wave, and eventually the acoustic wave amplifies. In turn the intense acoustic wave strongly interacts with pump wave and as a result the strength of scattered stokes mode enhances substantially.



Fig. 2. Variation of $(\chi_r^{(2)}(\omega_s))_{\beta}$ with magnetostatic field (B_0) and free carrier concentration (n_0) .

By selecting appropriate value of B_0 (say 9.0 T) and n_0 (say 1.5 x 10¹⁹ m⁻³) we obtained $(\chi_r^{(2)}(\omega_s))_\beta \sim 3.7$ x 10⁻⁹ (SI unit). When we varied both B_0 and n_0

simultaneously, the change in sign of $(\chi_r^{(2)}(\omega_s))_\beta$ is observed between 0.0 T to 10.5 T. Besides, it is also noticed that the value of $(\chi_r^{(2)}(\omega_s))_\beta$ varies almost parabolically with rising n₀. This figure also exhibits, when Ω_p (through n₀) approaches to $2\omega_a$, the frequency of coherent scattered stokes mode shift towards higher value of B₀ (i.e. blue shift). Interestingly, with further increase of Ω_p , due to departure from resonance, the coherent scattered stokes mode shift towards lower value of magnetic field (i.e. red shift).



Fig. 3. Variation of $(\chi_{r}^{(2)}(\omega_{s}))_{\beta,I}$ with magnetostatic field (B_{0}) and free carrier concentration (n_{0}) .

For 10.5 T < B₀ <14.0 T, the sign of $(\chi_r^{(2)}(\omega_s))_\beta$ is negative and its value is comparatively small. At B₀=14.1 T, the sign of $(\chi_r^{(2)}(\omega_s))_\beta$ changes due to the second resonance condition and the magnitude of $(\chi_r^{(2)}(\omega_s))_\beta$ is ~3.4 x 10⁻⁷ (SI Unit) and which is 10 times larger than the reported value in GaAlAs [34]. In particular, this result indicates that in the presence of strong externally applied magnetic field (~14.1T), the effect of free carrier concentration on $(\chi_r^{(2)}(\omega_s))_\beta$ is weak since $\Omega_p \square \omega_c$. However, in the absence of external magnetostatic field (B₀=0) and n₀= 1.5 x10¹⁹ m⁻³, the value of $(\chi_r^{(2)}(\omega_s))_\beta$ is 3.1 x 10⁻¹¹ (SI unit) which is much smaller than the value obtained around above mentioned resonance conditions. At B₀=0.0T, the pump wave scatters from acoustic wave and plasmon density in doped semiconductors.

The salient feature of result is the tailoring $(\chi_r^{(2)}(\omega_s))_\beta$ by varying independently/simultaneously B_0 and n_0 and also achieving large value of $(\chi_r^{(2)}(\omega_s))_\beta$ in III-V weakly piezoelectric semiconductors. The result allows tuning of strong coherent backward Stokes wave over a wide frequency range and opens up possibility of fabrication of frequency converters.

Fig 3 shows variation of the real part of intensitydependent-second-order susceptibility $(\chi_r^{(2)}(\omega_s))_{\beta I}$ (second term of eq. 13) with respect to B_0 and n_0 . This figure also depicts the enhancement of $(\chi_r^{(2)}(\omega_s))_{\beta,I}$ as well as change in its sign. However, in this case typical behavior of $(\chi_r^{(2)}(\omega_s))_{\beta I}$ arises due to three resonance conditions: (i) $\omega_s^2 = (\Omega_p^2 \omega_c^2 + v^2)/v^2$, (ii) $\omega_s^2 = 2\omega_c^2$ and (iii) $\omega_s^2 = \omega_c^2$. Interesting feature of this case is the interaction between plasmon oscillator and electroncyclotron oscillator. Let us define this oscillator as hybridoscillator. The hybrid-oscillator interacts with electric field of pump wave and as a result plasma and electroncyclotron frequencies dependent coherent scattered stokes wave is produced. It is often advantages, moving the frequency of coherent stokes wave to higher and more accessible spectral region in proportion to: (a) plasma frequency (or n_0) for fixed electron-cyclotron frequency (or B_0), (b) electron-cyclotron frequency for fixed plasma frequency and (c) combination of both frequencies. For example, in moderately doped semiconductor $(n_0=1.5 \times 10^{19} \text{m}^{-3})$, low externally applied magnetostatic field (1.5 -3.5 T) yields a large value of $(\chi_r^{(2)}(\omega_s))_{\beta I}$ and also alters its sign as well as shifts the stokes frequency towards higher frequency (Blue shift).

One may notice that between 4.0 to 9.0 T, the sign of $(\chi_r^{(2)}(\omega_s))_{\beta,I}$ is negative and its value is almost constant because in this region the resonance condition does not occurs. However, between 9.5 to 14.1 T, second $(\omega_s^2 \Box 2\omega_c^2)$ and third $(\omega_s^2 \Box \omega_c^2)$ resonance condition yield, sharp enhancement and change of sign of $(\chi_r^{(2)}(\omega_s))_{\beta,I}$ at B₀=10.0 T and 14.1 T, respectively.

Here it is worth pointing out that in case of $(\chi_r^{(2)}(\omega_s))_{\beta I}$ apart from B₀ and n₀ dependence, intensity dependence can also be utilized for the enhancement of $(\chi_r^{(2)}(\omega_s))_{\beta I}$. The value of $(\chi_r^{(2)}(\omega_s))_{\beta I}$ can be increased linearly with increasing pump intensity for a given value of B_0 and n_0 . At certain pump intensity I_p , the magnitude of $(\chi_{r}^{(2)}(\omega_{s}))_{\beta,I}$ is comparable to the value of $(\chi_r^{(2)}(\omega_s))_{\beta}$. For example when $B_0 = 0.0$ T, $(\chi_{r}^{(2)}(\omega_{s}))_{\beta,I} \sim (\chi_{r}^{(2)}(\omega_{s}))_{\beta}$ at excitation pump intensity $I_p \sim 1.6 \times 10^{12} \text{ W/m}^2$. Here it is worth mentioning that practically the excitation intensity can not be increased arbitrarily because it may damage the sample. Mayer et al [35] pointed out that when semiconductor is irradiated with intense laser light with long pulse duration a frequent consequence is the production of heat. However, the damage threshold can be lowered by reducing the pulse duration or through free carrier nonlinear absorption mechanism.

In fig 4 and 5 the imaginary part of eq. (13) has been analyzed. Fig 4 shows the variation of $(\chi_i^{(2)}(\omega_s))_\beta$ (imaginary part of first term) with magnetostatic field at two different free carrier concentrations 1.5×10^{19} m⁻³ and 2.4×10^{19} m⁻³. For both concentrations initially the $(\chi_i^{(2)}(\omega_s))_\beta$ is positive and grows very slowly with increasing B_0 . When $\omega_s^2 = (\omega_c^2 + v^2)/(1 - (\Omega_p^2/4\omega_a^2))$, the $(\chi_i^{(2)}(\omega_s))_\beta$ rises abruptly and reaches to its peak value and change of sign occurs at sharp resonance. Further negative $(\chi_i^{(2)}(\omega_s))_\beta$ decreases sharply due to departure from resonance and attains small value. On further increase in magnetostatic field an abrupt rise in $(\chi_i^{(2)}(\omega_s))_{\beta}$ and its change of sign is observed due to resonance condition $\omega_s^2 = \omega_c^2$.



Fig. 4. Variation of $(\chi_i^{(2)}(\omega_s))_{\beta}$ with magnetostatic field at free carrier concentrations (a) $n_0=1.5x10^{19} \text{ m}^{-3}$ and (b) $n_0=2.4x10^{19} \text{ m}^{-3}$.



Fig. 5. Variation of $(\chi_i^{(2)}(\mathcal{O}_s))_{\beta,I}$ with magnetostatic field at free carrier concentrations (a) $n_0 = 1.5 \times 10^{19} \, \text{m}^{-3}$ and (b) $n_0 = 2.4 \times 10^{19} \, \text{m}^{-3}$.

Above results infer when $\Omega_{\rm p} \sim 2\omega_{\rm a}$, reversal of sign of $(\chi_{\rm i}^{(2)}(\omega_{\rm s}))_{\beta}$ can be achieved in low doped piezoelectric semiconductor by applying low magnetic field. Hence, externally applied magnetostatic field dependent behavior of $(\chi_{\rm i}^{(2)}(\omega_{\rm s}))_{\beta}$ of moderately doped piezoelectric semiconductor sample may be utilized to design parametric switch (gain/loss) in the far infrared regime because positive/negative $(\chi_{\rm i}^{(2)}(\omega_{\rm s}))_{\beta}$ leads to gain/loss phenomenon.

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Fig. 5 shows the variation of imaginary part of second term with magnetostatic field at two different free carrier concentrations 1.5x10¹⁹ m⁻³ and 2.4x10¹⁹ m⁻³. For both concentrations, initially $(\chi_i^{(2)}(\omega_s))_{\beta,I}$ is positive and grows very slowly with increasing B₀. When electron cyclotron frequency approaches to stokes frequency, $(\chi_i^{(2)}(\omega_s))_{\beta,I}$ rises abruptly and reaches to its peak value. A change of sign occurs at sharp resonance condition $\omega_s^2 = (\Omega_p^2 \omega_c^2 + v^2) / v^2$ and saturate to small value due to departure from the resonance. Further increase in magnetostatic field gives rise to sharp enhancement of $(\chi_i^{(2)}(\omega_s))_{\beta,I}$ and changes of sign due to resonance condition $\omega_s^2 \sim 2\omega_c^2$. This figure also shows that the first resonance condition shifts toward lower magnetostatic field with increase in carrier concentration, while the second resonance condition (around $B_0=10.0$ T) is independent of free carrier concentration. For example for $n_0 = 1.5 \times 10^{19} \text{ m}^{-3}$ resonance occurs at 2.9T and it shifts to lower magnetostatic field ($B_0 = 2.3T$) when $n_0 = 2.4x$ 10^{19} m^{-3} .

4. Conclusions

In the present paper, using electromagnetic treatment the effective second-order optical susceptibility has been studied in weakly piezoelectric narrow direct-gap III–V doped semiconductors like n-InSb subjected to a transverse magnetostatic field under off-resonant transition regime. The following important conclusions may be drawn:

1. The hydrodynamic model of semiconductor-plasma has been successfully applied to study the effect of doping level, magnetostatic field and pump intensity on secondorder optical susceptibility in piezoelectric III–V doped semiconducting crystals duly illuminated by slightly offresonant not-too-high-power pulsed lasers with pulse duration sufficiently larger than the acoustic phonon lifetime. Moreover, the present analysis appears to be the first of its kind with detailed incorporation of convective nonlinearity and explored the possibility of intensity dependence of second-order optical susceptibility in III-V semiconductors.

2. Resonance between (i) lattice frequency and plasma frequency (ii) stokes frequency and electron-cyclotron frequency (iii) stokes frequency and hybrid frequency can

lead to large tunable effects specifically related to scattering phenomena and frequency tuning which can be utilized for spectroscopic measurement and frequency converters.

3. Near resonance conditions, significant enhancement of $\chi^{(2)}(\omega_s)$ as well as change in its sign can be achieved in weakly piezoelectric doped III-V semiconductors. This study may be utilized in observation of different phenomena such as self focusing/defocusing and absorption/gain. A strong transverse magnetostatic field (14.1T) enhances the $\chi^{(2)}(\omega_s)$ by a factor of 10⁴ as in its

absence.

4. By proper selection of doping concentration, externally applied magnetostatic field and intensity of pump, the value of second-order optical susceptibility in III-V semiconductors can be substantially enhanced and its behavior can also be tailored. The proposed model opens up possibilities for the fabrication of efficient and fast nonlinear optical devices.

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