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Original Research Article

Estimation of quadratic and cubic optical nonlinearities of transversely magnetized semiconductors

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ABSTRACT
Using coupled mode approach, an expression is obtained for the effective nonlinear optical susceptibility of transversely magnetized (TM) semiconductors under the off-resonant optical transition regime. The origination of laser-semiconductor interaction is considered in the nonlinear polarization arising via piezoelectric and electrostrictive properties of TM semiconductor. For numerical analysis, we consider the illumination of n-type doped indium antimonide sample by a pulsed carbon di-oxide laser at 1.78×10^{14} s^{-1} frequency. The authors make the effort for optimizing the doping concentration of the chosen sample and TM field to achieve enhanced values of quadratic and cubic optical nonlinearities of the chosen nonlinear medium. The authors found good agreement between their calculated values and the experimental observed values of quadratic and cubic optical nonlinearities. The analysis establishes that the quadratic and cubic optical nonlinearities of TM semiconductors can be increased by the significant amount by properly selecting the doping level and TM field, which proves the potential of chosen nonlinear medium for the fabrication of efficient and fast responding optoelectronic devices.

1. Introduction

In nonlinear optics, the enhancement of quadratic and cubic optical nonlinearities has been an interesting subject matter of research owing to their importance in fabrication of optoelectronic devices. The knowledge of quadratic and cubic optical nonlinearities provides necessary significant information regarding nonlinear optical characteristics of the nonlinear media [1, 2]. Among various existing and fabricated nonlinear optical materials, the elemental as well as compound semiconductors offer enormous litheness in the design and fabrication of modern optoelectronic devices. It is because:

(i) the carrier's recombination time of elemental as well as compound semiconductors can be controlled via fabrication and device structuring;

(ii) the change in either coefficient of absorption and/or index of refraction of elemental as well as compound semiconductor may be used for device fabrication;

(iii) the elemental as well as compound semiconductor based optoelectronic devices may be operated under the normal/ oblique incidence as well as in waveguides;

(iv) the elemental as well as compound semiconductors based optoelectronic devices use conventional semiconductor technology; and

(v) the elemental as well as compound semiconductors based optoelectronic devices can be easily integrated with other optoelectronic components.

Till date, the quadratic and cubic optical nonlinearities in the resonant transition regime, owing to their large values, have been used to enhance the efficiency of conversion of optoelectronic devices [3]. But these nonlinearities suffer from the serious drawback. The operating speed of the resonant quadratic and cubic optical nonlinearities based optoelectronic devices are very slow because the resonant quadratic and cubic optical nonlinearities are strongly dependent on the change in population incurred during the real transitions, buildup of photon energy and the time of relaxations. On the other hand, the non-resonant quadratic and cubic optical nonlinearities are, in general, comparatively smaller than the resonant quadratic and cubic optical nonlinearities but demonstrate much faster responses, because these does not entail the creation and relaxation of charge carriers.

In the future high speed optical communication systems, the non-resonant quadratic and cubic optical nonlinearities of compound semiconductors have found technologically important. Therefore, looking at the potential of non-resonant quadratic and cubic optical nonlinearities in future fast optical communications and to get better the performance of modern optoelectronic devices, there is a great need to enhance the non-resonant quadratic and cubic optical nonlinearities of compound semiconductors. Literature survey reveals that the doping (free carrier concentration), compassioning, and micro-level structuring techniques have been generally used to improve the efficiency and other parameters of compound semiconductor based devices in today's optoelectronic device technology [4 - 6].

The quadratic and cubic optical nonlinearities of compound semiconductors can be tailored by the application of

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an external TM field. This technique has been used in deep understanding the mechanisms included in various nonlinear processes including electro/magneto-optic effects. In the present paper, we develop a mathematical model to obtain expressions for effective quadratic and cubic optical susceptibilities ($\chi_e^{(2)}$ and $\chi_e^{(3)}$) of TM semiconductors.

2. Objectives

- To find the suitable values of carrier's concentration and applied TM field for enhancing the magnitude of $\chi_e^{(2)}$ and $\chi_e^{(3)}$ and change of their sign.
- To establish the potential of TM semiconductors for the fabrication of modern optoelectronic devices.

3. The Model

Let us consider the illumination of a compound semiconductor by an intense pump (laser) beam $\vec{E}_1(\omega_1, k_1\hat{z})$ such that the photon energy ($\hbar\omega_1$) of pump wave is slightly less than the band-gap energy ($\hbar\omega_g$) of the chosen compound semiconductor. Under this (off-resonant laser irradiation), the optical characteristics of the chosen compound semiconductor are affected by the doping level and remains un-affected by the photon-induced inter/ intra-band transitions. The force exerted by the pump wave on semiconductor is the cause of nonlinearity. The present mathematical formulation is carried out under the hydrodynamic model of semiconductor-plasmas. In this model, the wavelength linked with the lattice vibrations is sufficiently greater than the distance between the lattice points of the semiconductor crystal.

The (high frequency) oscillating pump field induced the electrostrictive and the piezoelectric strains and is thus derive an acoustic wave in the TM semiconductor. Under the influence of these strains, let the lattice point of semiconductor crystal gets deviated from its equilibrium coordinate z through a distance $u(z,t)$. Thus, the strain developed in the pump wave propagation direction may be expressed as: $\partial u/\partial z$. The effective force acting in the pump wave propagation (along +z) direction per unit volume of the TM semiconductor crystal can be expressed as:

$$f_{eff} = \frac{\partial}{\partial z}\left[-\beta(E_1) + 0.5\gamma|E_1|^2\right], \tag{1}$$

where β is electrostriction coefficient and γ is piezoelectric coefficient of chosen TM semiconductor. The equation of motion for $u(z,t)$ may be expressed as:

$$\rho\frac{\partial^2 u}{\partial t^2} - C\frac{\partial^2 u}{\partial z^2} + 2\Gamma\frac{\partial u}{\partial t} = \hat{z}f_{eff}, \tag{2}$$

where ρ, C, and Γ stand for the semiconductor crystal density, semiconductor elastic coefficient and the damping coefficient (introduced phenomenologically) of chosen semiconductor, respectively. Here, it is assumed that the acoustic wave set up in the TM semiconductor crystal is a travelling plane wave and defined as:

$$\vec{E}_2 = E_2\exp[i(\omega_2 t - k_2\hat{z})]. \tag{3}$$

The acoustical wave set up in the chosen TM semiconductor causes the scattering of the pump wave (at ω_3) that corresponds to the frequency of the material excitations, which, consequently causes the modulation of relative permittivity of the TM semiconductor. This leads to an exchange of photon energy among the electromagnetic radiations differing in frequency by an amount equal to the frequency of generated acoustic wave. Here, it should be noted that, the scattered wave is also a travelling plane wave and it may be defined as: $\vec{E}_3[= E_3\exp i(\omega_3 t - k_3\hat{z})]$.

Here, it should be mentioning that the pump beam is an infrared pulsed laser having frequency $\omega_1 >> \omega_2$ with pulse time duration $t_p > \Gamma^{-1}$. This condition allows treating the laser-semiconductor interaction as a quasi steady-state interaction. In the presence of optical and TM fields, the equations describing the motion of doped (free) carriers may be expressed via 0th and 1st order momentary transfer equations as:

$$\frac{d\vec{v}_0}{dt} + \nu\vec{v}_0 = -\frac{e}{m}\vec{E}_e \tag{4a}$$

$$\frac{d\vec{v}_1}{dt} + \nu\vec{v}_1 + \vec{v}_1\frac{d\vec{v}_0}{dz} = -\frac{e}{m}[\vec{E}_1 + \vec{v}_1\times B_q]. \tag{4b}$$

In Eqs. (4a) and (4b), the parameters v_0 and v_1 stand for the steady-state and perturbed fluid velocities of doped carrier (viz. an electron) of effective mass m and charge $-e$, respectively. ν represents the electron collision frequency. $\vec{E}_e\left[= \vec{E}_1 + \vec{v}_0\times\vec{B}_q\right]$ is the net electric field; including the magnetic Lorentz's force ($\vec{v}_0\times\vec{B}_q$). At optical frequencies, the net field exerts force on the doped (electron) carrier's because of their small effective mass.

The needful equations employed in the derivation of quadratic and cubic optical susceptibilities of TM semiconductors are:

$$\frac{\partial n_1}{\partial t} + n_0\frac{\partial v_1}{\partial z} + n_1\frac{\partial v_0}{\partial z} + v_0\frac{\partial n_1}{\partial z} = 0 \tag{5}$$

$$\frac{\partial E_{sc}}{\partial z} = -\frac{1}{\varepsilon}\left[n_1 e + \beta\frac{\partial^2 u}{\partial z^2} - \gamma\frac{\partial^2 u^*}{\partial z^2}E_1\right]. \tag{6}$$

Eq. (5) is continuity equation. Here, n_0 is the equilibrium doped carrier's density, and n_1 is the perturbed doped carrier's density. Eq. (6) is the Poisson's equation and it determines the space-charge electric field E_{sc} originating via electrostrictive and piezoelectric strains and via density perturbations in the TM semiconductor. Other quantities have their usual meanings.

The nonlinear interaction between the charge carriers (electrons) and the acoustic wave set up in the TM semiconductor is the origin of optical nonlinearity. Further, we consider that $\omega_c\tau << 1$, where the charge carrier does not have much chance of being deflected by the pump field before undergoing a collision, this distance is the carriers mean free path l. On the other hand, under the influence of strong TM field, $\omega_c\tau >> 1$, the charge carriers are able to make several

revolutions in their orbits before their scattering occurs and this distance ~ cyclotron radius. In compound TM semiconductors, the equation of carrier fluctuations can be derived from Eqs. (1) - (6) as:

$$\frac{\partial^2 n_1}{\partial t^2}+\nu\frac{\partial n_1}{\partial t}+\omega_p^2\delta_1 n_1 - i\frac{k^3\omega_p^2\delta_1}{2e\rho\omega_D^2}\left[\beta\gamma E_1 E_3^* - \gamma^2\left|E_1\right|^2 E_3\right] = (e/m)\delta_0 E_1\left(\frac{\partial n_1}{\partial z}+ik_1 n_1\right) \quad (7)$$

where

$\omega_c = -\frac{e}{m}B_q$ (electron cyclotron frequency),

$\omega_p = \sqrt{\frac{n_0 e^2}{m\varepsilon}}$ (electron-plasma frequency),

$\delta_0 = 1-\frac{\omega_c^2}{\Delta_0^2+\omega_c^2}$, $\delta_1 = 1-\frac{\omega_c^2}{\Delta_1^2+\omega_c^2}$,

$\Delta_0 = \nu - i\omega_1$, and $\Delta_1 = \nu + i(\omega_3 - ik_3 v_1)$,

$v_2[=(C/\rho)^{1/2}]$ is the velocity of acoustic phonon mode in the TM semiconductor, and

$\omega_D^2(\omega,k)[=(\omega_2^2 - k_2^2 v_2^2 + 2i\Gamma\omega_2]$ stands for the acoustic-wave dispersion.

While deriving Eq. (7), we neglected the Doppler's shift under the condition $\omega_1 >> k_2 v_2$. For pump energy very much less than the band gap energy of TM semiconductor i.e. $\hbar\omega_1 << \hbar\omega_g$, the scattered anti-Stokes field can be neglected and only the scattered Stokes field is considered [3]. The resonant scattered Stokes component may be obtained by using the phase matching constraints: $\omega_3 = \omega_1 - \omega_2$ and $\vec{k}_3 = \vec{k}_1 - \vec{k}_2$.

The perturbed carrier's concentration n_1 consists of two frequency components viz., slow frequency component of carrier concentration (n_{1s}) and fast component of carrier concentration (n_{1f}). The slow frequency component of carrier concentration oscillates at the acoustic phonon mode frequency while the fast component of carrier concentration oscillates at Stokes mode frequency. In the present mathematical modeling, the resonant sideband frequencies ($\omega_1 \pm q\omega_2$), (with q = 1, 2, 3, …) are considered under the assumption that the interaction path of carriers is sufficiently long. Moreover, the higher-order Stokes components (for $q \geq 2$) of resonant side band frequencies have been neglected under off-resonant laser irradiation and only the first-order resonant Stokes component (with $q = 1$) has been taken into account. Under RWA, Eq. (7) may be split into to the following coupled mode equations:

$$\frac{\partial^2 n_{1s}}{\partial t^2}+\nu\frac{\partial n_{1s}}{\partial t}+\omega_p^2\delta_1 n_{1s}+i\frac{k^3\omega_p^2\delta_1}{2\rho e\omega_D^2}E_1 E_3^* = i\frac{e}{m}k\delta_0 E_1 n_{1s}^* \quad (8a)$$

$$\frac{\partial^2 n_{1f}}{\partial t^2}+\nu\frac{\partial n_{1f}}{\partial t}+\omega_p^2\delta_1 n_{1f}+i\frac{k^3\omega_p^2\delta_1\gamma^2}{2\rho e\omega_D^2}\left|E_1\right|^2 E_3 = i\frac{e}{m}k\delta_0 E_1 n_{1s}^*. \quad (8b)$$

Eqs. (8a, b) demonstrate that n_{1s} and n_{1f} of perturbed carrier concentration are coupled via the pump field. The expression for n_{1s}^* may be obtained as:

$$n_{1s}^* = \frac{i\omega_p^2 k^4\delta_0\delta_1}{2\rho m\omega_D^2\Omega_0^2\Omega_1^2}\left[\beta\gamma E_1 - \gamma^2\left|E_1\right|^2\right]E_1 E_3^*, \quad (9)$$

where $\Omega_0^2 = (\omega_p^2\delta_0 - \omega_2^2 - i\nu\omega_2)$
and $\Omega_1^2 = (\omega_p^2\delta_1 - \omega_3^2 - i\nu\omega_3)$.

The resonant stokes component of the effective value of nonlinear current density arising via perturbed carrier density may be derived by using the equation:

$$J_e(\omega_3,k_3) = n_0 e v_{1s} + n_{1s} e v_1. \quad (10)$$

In coupled-mode theory of interacting waves, the time integral of $J_e(\omega_3,k_3)$ gives the effective value of nonlinear polarization as:

$$P_e = \int J_e(\omega_3,k_3)dt. \quad (11)$$

Using Eqs. (9) – (11), the effective value of nonlinear polarization may be obtained as:

$$P_e = \frac{\varepsilon_0\varepsilon_l\omega_p^2 E_3^*}{\omega_3(i\nu+\omega_3)} - i\left[\frac{\omega_p^2 k^3\delta_0\delta_1(e/m)^2}{2\rho\omega_D^2\omega_s^2\Omega_0^2\Omega_1^2}\right]\left[\beta\gamma E_1 E_3^* - \left(\beta\gamma - \gamma^2(m/ek_2)\omega_1^2\right)\left|E_1\right|^2 E_3^*\right] \quad (12)$$

Using Eq. (12), and the generalized equation describing the effective value of nonlinear polarization viz., $P_e = \varepsilon_0\left[\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + ...\right]$, the various orders (viz. first, second, third, …) of optical susceptibilities of TM semiconductors can be obtained as:

$$\chi^{(1)}[=\chi_r^{(1)} + i\chi_i^{(1)}] = \frac{\varepsilon_l\omega_p^2}{\omega_3(i\nu+\omega_3)}, \quad (13)$$

$$\chi_e^{(2)}[=\chi_r^{(2)} + i\chi_i^{(2)}] = -i\frac{\omega_p^2 k^3\beta\gamma\delta_0\delta_1(e/m)^2}{2\rho\varepsilon_0\omega_D^2\omega_s^2\Omega_0^2\Omega_1^2} \quad (14)$$

$$\chi_e^{(3)}[=\chi_r^{(3)} + i\chi_i^{(3)}] = i\frac{\omega_p^2 k^3\delta_0\delta_1(e/m)^2}{2\rho\varepsilon_0\omega_D^2\omega_s^2\Omega_0^2\Omega_1^2}\left[\beta\gamma - \gamma^2(m/ek_2)\omega_1^2\right] \quad (15)$$

Eqs. (14) and (15) reveal that that the origin of quadratic optical susceptibility $\chi_e^{(2)}$ lies in the finiteness of both the electrostrictive coefficient γ and the piezoelectric coefficient β. On the other hand, the origin of cubic optical susceptibility $\chi_e^{(3)}$ lies in finiteness of the electrostrictive coefficient γ while the piezoelectric coefficient β adds new dimension.

Eqs. (13) – (15) also reveal that the linear, quadratic, and cubic optical susceptibilities, all are complex quantities. The imaginary and real parts of $\chi_e^{(1)}$ account for the linear absorption and index of refraction. The quadratic optical susceptibility $\chi_e^{(2)}$ gives rise the 2nd order optical effects. The cubic optical susceptibility $\chi_e^{(3)}$ gives rise the 3rd order optical effects.

4. Results and discussion

For the numerical estimation of quadratic and the cubic optical susceptibilities ($\chi_e^{(2)}$ and $\chi_e^{(3)}$), the authors considered the irradiation of an n-type doped indium antimonide crystal by a pulsed carbon di-oxide laser. The physical parameters of the chosen laser-semiconductor interaction system are given in the Ref. [7]. Fig. 1 depicts the 3D variation of real part of $\chi_e^{(2)}$ and $\chi_e^{(3)}$ with TM field B_q and doping concentration n_0.

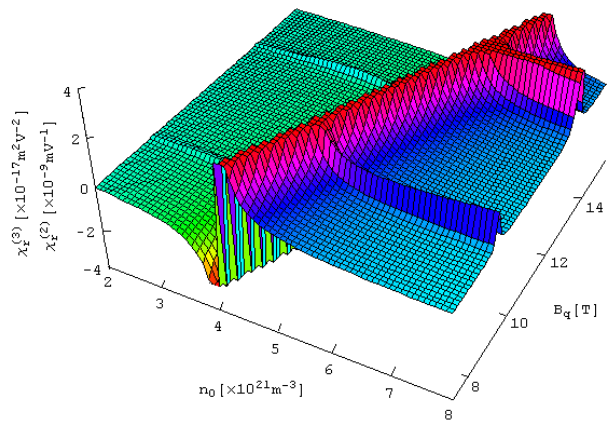

Figure 1. 3D variation of $\chi_r^{(2)}$ and $\chi_r^{(3)}$ with n_0 and B_q.

It can be seen that the TM field significantly modifies the real part of quadratic and cubic optical susceptibilities ($\chi_r^{(2)}$ and $\chi_r^{(3)}$). Both $\chi_r^{(2,3)}$ exhibit sharp peak as well as change of sign (from negative to positive) exactly at TM field $B_q = 10$ and 14 T because of the resonance conditions $2\omega_c^2 \sim \omega_0^2$ and $\omega_c^2 \sim \omega_0^2$, respectively. Thus around resonances, the Lorentz contribution of TM field is very effective in enhancing $\chi_r^{(2,3)}$ in TM semiconductors. However, in the regime $10 \le B_q \le 14.2$ T, $\chi_r^{(2,3)}$ are fairly independent of B_q. Thus around off-resonances ($2\omega_c^2 \le \omega_l^2 \le \omega_c^2$), the Lorentz contribution of TM field is absent. With further increasing TM field B_q, $\chi_r^{(2,3)}$ drops quickly because of the departure from resonances and saturating at a comparatively smaller value. Also, the doping concentration imparts significant role in modification of $\chi_r^{(2,3)}$.

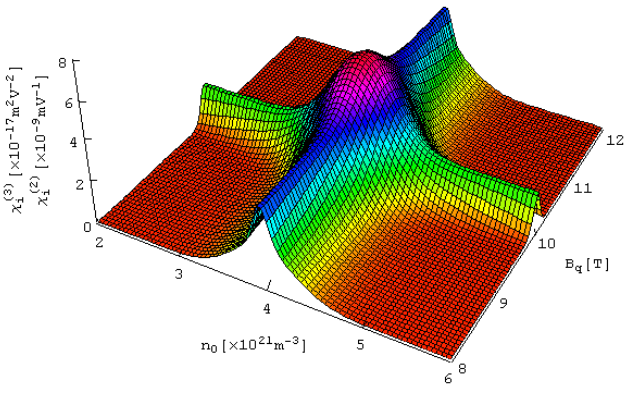

Figure 2. 3D variation of $\chi_i^{(2)}$ and $\chi_i^{(3)}$ with n_0 and B_q.

For $\omega_p < \omega_{3m}[= \omega_3(1-(2\omega_c^2/\omega_l^2))(1-(\omega_c^2/\omega_l^2))$, both $\chi_r^{(2,3)}$ are negative and falls rapidly with ω_p. With slightly making the tuning between ω_p and ω_{3m}, a sharp rise in both $\chi_r^{(2,3)}$ are observed. When ω_p is sharply tuned with ω_{3m}, both $\chi_r^{(2,3)}$ vanish. This critical TM field dependence of both $\chi_r^{(2,3)}$ can be used to fabricate the nonlinear optical switches. After resonances, both $\chi_r^{(2,3)}$ rise quite sharply and achieve maxima at $\omega_p = \omega_{3m} + \nu$.

Fig. 2 depicts the 3D variation of imaginary parts of $\chi_e^{(2)}$ and $\chi_e^{(3)}$ with TM field B_q and doping concentration n_0. Here, instead of change of sign, both $\chi_i^{(2,3)}$ exhibit peak around $\omega_p \sim \omega_{3m}$. Around resonance, the electron's drift speed, which is strongly dependent of TM field increases significantly and it becomes much greater than the acoustic wave speed. Consequently, the photon energy transfer from pump to acoustic wave increases, thereby enhancing $\chi_i^{(2,3)}$.

The calculated values of real and imaginary parts of $\chi_e^{(2)}$ and $\chi_e^{(3)}$ for the indium antimonide sample in the presence and absence of TM field are represented in Table 1.

Table 1. Calculated values of $\chi_i^{(2)}$, $\chi_r^{(2)}$, $\chi_i^{(3)}$ and $\chi_r^{(3)}$ for n_0 = 2×10^{22} m^{-3}.

Nonlinear optical susceptibilities	TM field		
	B_q = 0.0 T	B_q = 10.0 T	B_q = 14.2 T
$\chi_i^{(2)}$ (mV^{-1})	2.10×10^{-11}	3.92×10^{-9}	9.47×10^{-9}
$\chi_r^{(2)}$ (mV^{-1})	2.12×10^{-11}	4.20×10^{-9}	1.22×10^{-8}
$\chi_i^{(3)}$ (m^{2}V^{-2})	1.16×10^{-19}	3.90×10^{-17}	9.60×10^{-18}
$\chi_r^{(3)}$ (m^{2}V^{-2})	1.19×10^{-19}	4.18×10^{-17}	1.17×10^{-17}

5. Conclusions

In this paper, a numerical analysis is performed to estimate the quadratic and cubic nonlinear optical susceptibilities of TM semiconductors. The proper selection of TM field and doping concentration is found to enhance the quadratic and cubic optical nonlinearities of TM semiconductors as well as change of their sign. The mathematical model developed proves the potential of TM semiconductors for the fabrication of efficient and fat responding optoelectronic devices.

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