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Original Research Article

Nonlinear refractive index and absorption coefficient of scattered Stokes mode in n-type doped gallium arsenide

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ABSTRACT
Many scientists have become interested in the experimental and theoretical studies concerning the measurement of nonlinear optical susceptibilities in various media due to their potential applications in the development of more modern coherent laser sources over a wide frequency range, including parametric amplifiers and oscillators, optical switches, phase conjugate mirrors, and above all, parametric amplifiers and oscillators. I have looked into the nonlinear propagation of a high frequency pump radiation in an n-GaAs sample in the region of negative differential resistivity using a hydrodynamic model of plasma in this research. In order to help the pump wave move electrons from the lower conduction valley to the upper satellite valleys in n-GaAs and therefore increase the effective mass of electrons, a d.c. electric bias is introduced. The effective mass's energy dependence and the sample's piezoelectric properties lead to the nonlinearity. It is discovered that the induction of fourth-order nonlinearity in the sample is caused by the energy dependence of the mass in the region of negative differential resistivity. It is discovered that the nonlinear optical properties of the sample are significantly influenced by the negative differential resistivity area in n n-GaAs.

1. Introduction

Many scientists have become interested in the experimental and theoretical studies concerning the measurement of nonlinear optical susceptibilities in various media due to their potential applications in the creation of contemporary optoelectronic devices like parametric amplifiers and oscillators, optical switches, phase conjugate mirrors, and, most importantly, in the creation of newer coherent laser sources over a wide range of frequencies. This nonlinear crystal feature has been widely used for optical bistability [1], phase conjugation [2], adjusting infrared lasers, and other applications.

In crystalline media, the dominant optical processes may be characterized by second- or higher-order nonlinear optical susceptibilities $\chi^{(n)}(n \geq 2)$. In contrast to substances like gases and liquids, semiconductors have been the obvious choice as host media for the study of nonlinear optical phenomena due to the availability of sophisticated fabrication technology and the experimental observations of enormous optical nonlinearities in the band-gap resonant transitions [3–5]. Gallium arsenide (n-GaAs), which has a many-valley band structure, has an advantage over other semiconductor materials in the design of devices due to its NDR property and negative differential resistivity (NDR) property.

Study of nonlinear optical effects (refractive and absorptive) in semiconductors near the fundamental absorption edges is a new area of research that has recently been possible thanks to the availability of numerous steady laser sources in the infrared range [6, 7]. The nonlinear optical susceptibility of the crystal is connected to the nonlinear components of the refractive index and absorption coefficient. As a result, studies into nonlinear susceptibility are crucial, and numerous methods for comprehending the underlying mechanisms have previously been reported in the literature [8–10]. Authors have either attributed the first-order forces resulting from piezoelectricity, etc., or the second-order forces resulting from finite differential polarizability and/or electrostrictive strain as the cause of nonlinear optical effects in the works previously cited. But in GaAs, where a significant nonlinearity may develop through the field-dependent effective mass of an electron leading to NDR features, no such work has yet been documented.

It is well known that an n-GaAs sample exhibits negative differential resistivity (NDR) throughout a wide range of free-electron parameters when the d.c. electric field applied to the material exceeds a specific threshold limit. The intervalley scattering of conduction electrons from low-energy/high-mobility Γ-point satellite valleys to high-energy/low mobility X point satellite valleys is responsible for the NDR effects in n-GaAs. Here, we predict that an intense laser beam may result in intermittent electron transfers that reveal NDR effects in n-GaAs. Additionally, one may anticipate nonlinear interaction with acoustic disruption produced inside the medium due to the crystal's piezoelectricity or electrostriction property at such a high-power level of the laser beams. To our knowledge, no attempt has ever been made to ascertain the nonlinear optical susceptibility resulting from the field dependent effective mass of an electron, which in turn affects the nonlinear refractive index and effective absorption coefficient.

As a result, we have analytically examined the nonlinear refractive index and absorption coeficient for the scattered mode in n-GaAs in the current study. To help the laser beam induce population inversion in the valleys, a d.c. bias field is added to the specimen. Nonlinearity develops on a time scale shorter than the intervalley scattering period due to the piezoelectric pressure on electrons. The intervalley transmission of the electrons causes nonlinearity on the longer time scale. As a result, throughout the entire range of time scales, one may think of piezoelectricity and intermittent electron transport as nonlinearity causes. Piezoelectricity initiates interaction, and as time passes, nonlinearities resulting from the intermittent flow of electrons take control.

2. Theoretical formulations

This section deals with the theoretical formulation of the total induced current density $\vec{J}$ for the signal and Stoke's component of scattered electromagnetic wave in n-GaAs crystal. We have considered the well-known hydrodynamic model of plasma subjected to an electromagnetic field. In order to study the total induced current density $\vec{J}$, we consider the propagation of a high frequency pump radiation along the *x*-direction in an n-GaAs sample biased with a d.c. electric field in the *x*-direction.

$$\vec{E}_0[E_{0x}\hat{x}+E_{0y}\hat{y}]\exp[i(k_0x-\omega_0t)]\,. \tag{1}$$

The other basic equations considered for the analysis are:

$$\frac{\partial^2 u}{\partial t}+\frac{C}{\rho}\frac{\partial^2 u}{\partial x^2}+2\Gamma_a\frac{\partial u}{\partial t}=-\frac{\beta}{\rho}\frac{\partial E_a}{\partial x} \tag{2}$$

$$\frac{\partial \vec{v}_0}{\partial t}+\nu\vec{v}_0=-\frac{e}{m}\vec{E}_0 \tag{3}$$

$$\frac{\partial \vec{v}_1}{\partial t}+\nu\vec{v}_1+v_{0x}\frac{\partial \vec{v}_1}{\partial x}=-\frac{e}{m}\vec{E}_1 \tag{4}$$

$$\frac{\partial n_1}{\partial t}+n_e\frac{\partial v_1}{\partial x}+n_1\frac{\partial v_0}{\partial x}+v_0\frac{\partial n_1}{\partial x}=0 \tag{5}$$

$$\frac{\partial E_a}{\partial x}=-\frac{n_1e}{\varepsilon}-\frac{\beta}{\varepsilon}\frac{\partial^2 u}{\partial x^2}\,. \tag{6}$$

Equation (2) represents the motion of lattice in the crystal. Hence $\vec{u}(x,t)=\vec{u}\exp[i(k_ax-\omega_at)]$ denote the relative displacement of oscillators from the mean position of the lattice. *C* is the linear elastic modulus, β is the piezoelectric coefficient and ρ is the mass density of the crystal. $(\omega_a,\vec{k}_a)$ represents the frequency and wave vector of the generated acoustic phonon mode under consideration; Γ_a is the phenomenological damping parameter. Equations (3) and (4) represent the rate equations for the pump and the signal beam, $\vec{v}_0$, $\vec{v}_1$ are the oscillatory fluid velocities of the electrons of effective mass *m* and charge *-e*. ν is the electron collision frequency assumed to be a constant since its dependence on field is not very important at optical frequencies [11]. The electron continuity equation is given by Eq. (5) in which n_e and n_1 are the equilibrium and perturbed carrier densities, respectively. The space-charge field E_a is determined by the Poisson relation [Eq. (6)], where ε is the dielectric function of the semiconductor expressed equivalently to $\varepsilon_0\varepsilon_L$; ε_0 and ε_L are the absolute permittivity and relative dielectric constant of the crystal, respectively. In the above relations, we have neglected the effect due to $\vec{v}_0\times\vec{B}_1$ by assuming that the propagating acoustic mode is producing a longitudinal electric field.

Following Ref. [12, 13], the effective mass m of electrons (over a time scale greater than the intervalley scattering time τ_1, i.e. $\omega(\tau_1^{-1})$ is given by

$$m=m(E_T^2)$$
$$=m_0+m_2\left[\vec{E}_d.\vec{E}+\frac{\nu\omega_a}{2\omega_0}\left(\frac{\vec{E}_0.\vec{E}_s}{\omega_s}-\frac{\vec{E}_0^*.\vec{E}_{as}}{\omega_{as}}\right)\right], \tag{7}$$

where $m_0=m(E_{eff}^2)$, $m_2=\dfrac{\partial m}{\partial E_{eff}^2}$, and $E_{eff}^2=\dfrac{E_d^2+\nu^2E_0^2}{\omega_0^2}$,

in which subscripts *T*, *s* and *as*" stand for total, Stoke's and anti-Stoke's components, respectively.

Following Ref. [13, 14], we note that the effective masses of electrons in n-GaAs are $0.072m_e$ in the central valley and $0.4m_e$ in each of the six satellite valleys, where m_e is the mass of an electron in free space. If we take the satellite valleys equivalent to a single valley, then the effective mass of an electron is $1.20m_e$. The energy difference between the central valley and the equivalent satellite valley is: $\Delta\varepsilon_{\Gamma x}=0.36$. At low temperature the electrons occupy the central valley. When an electric field, either a d.c. field or the field of a high frequency wave, is applied the electrons are heated and in collisions with the lattice transferred to the upper equivalent valley. We can obtain the variation of the average effective mass of an electron $m(E_{eff}^2)$ with E_{eff}^2 from the curves showing the variation of mobility and other transport coefficients [15, 16].

Now, applying the procedure adopted in earlier works [7, 9, 10] to the present field configuration, one obtains the following coupled equations under rotating wave approximation for perturbed electron concentrations n_f (fast component associated with $\omega_0\pm\omega_a$ and n_a (slow component associated with the low frequency acoustic wave ω_a):

$$\frac{\partial^2 n_f}{\partial t^2}+\nu\frac{\partial n_f}{\partial t}+\omega_p^2\left(1-\frac{m_2E_{eff}^2}{m_0}\right)n_f=-i(k_0-k_a)\bar{E}n_a^*, \tag{8a}$$

$$\frac{\partial^2 n_a}{\partial t^2}+\nu\frac{\partial n_a}{\partial t}+\omega_p^2\left(1-\frac{m_2E_{eff}^2}{m_0}\right)n_a+\frac{n_0e\beta}{m_0\varepsilon}\left(1-\frac{m_2E_{eff}^2}{m_0}\right)\frac{\partial^2 u}{\partial x^2}=-i(k_0-k_a)\bar{E}n_f^*, \tag{8b}$$

in which $\bar{E}=-(e/m)E_0$ and * denotes the complex conjugate of the quantity.

The above equations exhibit that the nonlinear polarization induced by the intense pump wave leads to the coupling of high-frequency electromagnetic waves and material excitations wave generated within the medium. The density perturbation associated with the acoustic phonon mode at frequency ω_a beats with the pump at frequency ω_0 and produces fast components of density perturbations leading to an exchange of energy using the electromagnetic fields separated in frequency by integral multiples of ω_a. Since we have neglected the higher harmonics, the Stoke's mode of the scattered component at $[\omega_s(=\omega_0-\omega_a),\vec{k}_s(=\vec{k}_0-\vec{k}_a)]$ can be obtained from Eqs. (2) and (8) as:

$$n_a=\frac{in_ee\beta^2k_a^3[1-(m_2/m_0)E_{eff}^2]E_a}{m_0\varepsilon\rho(\omega_a^2-k_a^2v_a^2+2i\Gamma_a\omega_a)}\times\left[\omega_p^2\left(1-\frac{m_2}{m_0}E_{eff}^2-\omega_a^2\right)-i\nu\omega_a-\frac{k_s^2\left|\bar{E}\right|^2}{\{\omega_p^2[1-(m_2/m_0)E_{eff}^2-\omega_s^2+i\nu\omega_s\}}\right]^{-1}. \tag{9}$$

The resonant Stoke's component of the induced current density due to finite nonlinear polarization of the medium has been deduced by neglecting the transition dipole moment, which can be represented as:

$$\begin{aligned}J_s(\omega_s)&=-n_s^*ev_0\\&=\frac{ie\beta^2k_a^3\omega_p^2[1-(m_2/m_0)E_{eff}^2]E_0E_a^*}{m_0\rho(\nu-i\omega_0)(\omega_a^2-k_a^2v_a^2-2i\Gamma_a\omega_a)}\times\left[\delta_a^2-\omega_p^2\frac{m_2E_{eff}^2}{m_0}+i\nu\omega_a-\frac{k_s^2\left|\bar{E}\right|^2}{\{\delta_s^2-\omega_p^2(m_2/m_0)E_{eff}^2-i\nu\omega_s\}}\right]^{-1},\end{aligned} \tag{10}$$

where $\delta_a^2=\omega_p^2-\omega_a^2$ and $\delta_s^2=\omega_p^2-\omega_s^2$.

Now treating the induced polarization $P_{cd}(\omega_s)$ as the the integral of Stoke's component of the induced current density $J_s(\omega_s)$, one may write

$$P_{cd}(\omega_s)=\int J_s(\omega_s)\,dt. \tag{11}$$

In order to study the total nonlinear optical susceptibility χ, we can express the induced polarization in a crystal in the form of an expansion series

$$P_{cd}(\omega_s)=\varepsilon_0[\chi^{(1)}+\chi^{(2)}\left|E_0\right|+\chi^{(3)}\left|E_0\right|^2++\chi^{(4)}\left|E_0\right|^3+.....]\bar{E}_a, \tag{12}$$

where $\chi^{(n)}$ are the n^{th} order susceptibilities due to induced current density. Thus one can find out different orders of susceptibility of the crystal by using Eqs. (11) and (12).

Using Eqs. (10) and (11), we will obtain an expression for $P_{cd}(\omega_s)$, and equating the expressions corresponding to the same power of $E_s(\omega_s)$ in this derived equation and Eq. (12), one finds

$$\chi_{cd}^{(2)}=-e\omega_p^2\beta^2k_a^3(\delta_s^2-i\nu\omega_s)\left[2\varepsilon_0m_0\rho\Gamma_a\omega_s\omega_a\omega_0\left(1-\left(\frac{k_s^2\left|\bar{E}\right|^2}{\delta_a^2+i\nu\omega_a}\left(\delta_s^2-i\nu\omega_s\right)\right)\right)\right]^{-1} \tag{13}$$

and

$$\chi_{cd}^{(4)}=-e\omega_p^2\beta^2k_a^3\nu^2(\delta_s^2-i\nu\omega_s)(m_2/m_0)\left[2\varepsilon_0m_0\rho\Gamma_a\omega_s\omega_a\omega_0\left(1-\left(\frac{k_s^2\left|\bar{E}\right|^2}{\delta_a^2+i\nu\omega_a}\left(\delta_s^2-i\nu\omega_s\right)\right)\right)\right]^{-1}. \tag{14}$$

From Eqs. (13) and (14), one can notice that the involved high-order nonlinearities in $\chi_{cd}^{(2)}$ and $\chi_{cd}^{(4)}$ are negligibly small in the weak to moderate excitation limits satisfying the condition:

$$\left(\frac{k_s^2\left|\bar{E}\right|^2}{\delta_a^2+i\nu\omega_a}\left(\delta_s^2-i\nu\omega_s\right)\right)<<1. \tag{15}$$

This consideration enables one to obtain the second order and fourth-order components of χ_{cd} from Eqs. (13) and (14) as:

$$\chi_{cd}^{(2)}=\frac{-e\omega_p^2\beta^2k_a^3(\delta_s^2-i\nu\omega_s)}{2\varepsilon_0m_0\rho\Gamma_a\omega_s\omega_a\omega_0}, \tag{16}$$

and

$$\chi_{cd}^{(4)} = \frac{-e\omega_p^2\beta^2 k_a^3 \nu^2 (\delta_s^2 - i\nu\omega_s)(m_2/m_0)}{2\varepsilon_0 m_0 \rho \Gamma_a \omega_s \omega_a \omega_0}. \tag{17}$$

From Eqs. (16) and (17), one notices that both $\chi_{cd}^{(2)}$ and $\chi_{cd}^{(4)}$ are complex in nature. The optical nonlinearities are explained by finite real and imaginary parts of $\chi_{cd}^{(2)}$ and $\chi_{cd}^{(4)}$ considering that the higher-order nonlinear susceptibilities contribute negligibly.

The most exciting result which we have obtained here is that a fourth order nonlinearity is introduced in the system due to field dependent mass (m_2/m_0) of GaAs. Thus the negative differential resistivity of GaAs attributes to the fourth order nonlinearity in the System.

Thus in an n-GaAs crystal the total optical susceptibility χ is given by

$$\chi = \chi_{cd}^{(2)} + \chi_{cd}^{(4)}. \tag{18}$$

From Eqs. (16) and (17), one may infer that the origin of $\chi_{cd}^{(2)}$ and $\chi_{cd}^{(4)}$ are piezoelectricity and the field dependency of electron mass in n-GaAs crystal. Second order optical parameters are studied in detail by many workers. Hence we will confine our discussion to fourth-order optical effects only.

From the real parts of the fourth order susceptibility, one can study the phenomenon of the nonlinear refractive index in n-GaAs crystal as they are given by [17]:

$$\eta^{(n)} = \mathrm{Re}[\chi^{(n)}], \tag{19}$$

where $\eta^{(n)}$ denotes the order of nonlinearity.

By substituting the imaginary part of $\chi_{cd}^{(4)}$ in the following relation, one can study the phenomenon of nonlinear absorption in n-GaAs crystal. The relation is:

$$\alpha_{nl}^{(n)} = -\frac{k_s}{2\varepsilon_L}.\mathrm{Im}[\chi^{(n)}]. \tag{20}$$

Now Eqs. (19) and (20) can be safely employed to study the nonlinear parameters (refractive and absorptive) of a scatand tered mode in n-GaAs plasma.

3. Results and discussion

This section is devoted to the detailed numerical study of the nonlinear refractive and absorptive characteristics of an n-GaAs plasma through fourth order nonlinear susceptibility arising due to the field dependent effective mass of electrons. The set of parameters appropriate for n-GaAs plasma, has been used in the numerical analysis are taken from Ref. [6]. The results are plotted in Figures 1-5.

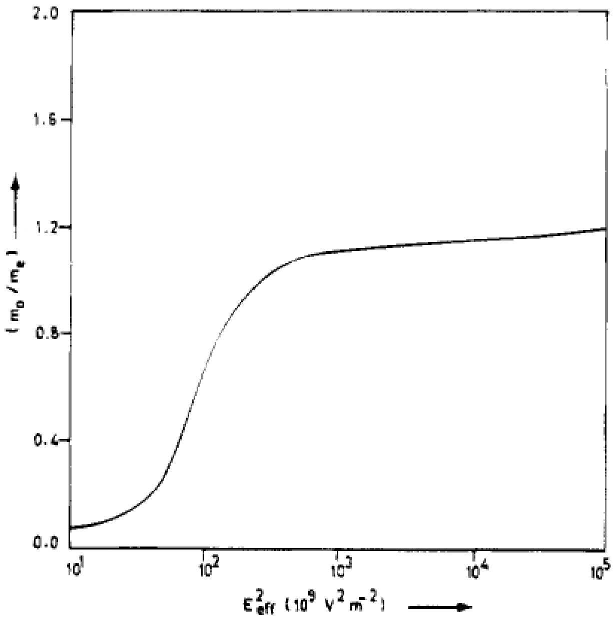

Figure 1: The variation of the average effective mass of electrons in n-GaAs, $m(E_{eff}^2)/m_e$ with E_{eff}^2.

Figure 1 shows the variation of the field-dependent average effective mass of an electron with E_{eff}^2. This curve is obtained from the probability distribution function of electrons in the Γ-point valley and the equivalent satellite valley, as given in Conwell [15]. For further calculations the values of m_0 and m_2 have been taken from Figure 1.

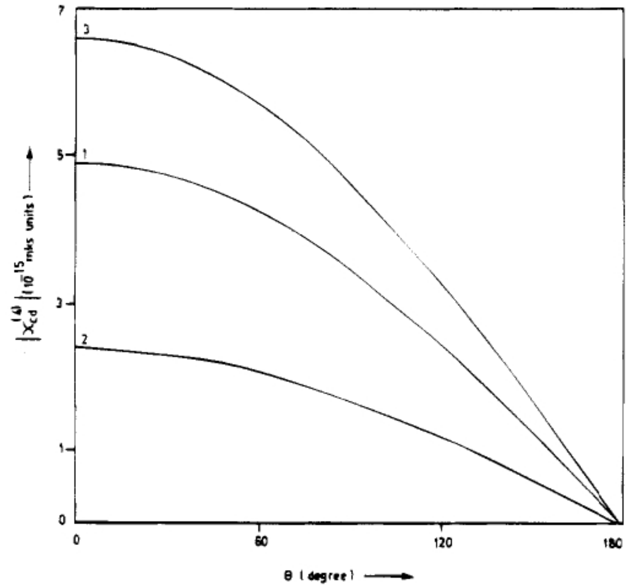

Figure 2: The variation of the magnitude of $\chi_{cd}^{(4)}$ with the scattering angle θ for $E_d = 1.5\times10^3$ Vm^{-1}. The curve 1 corresponds to $(m_2/m_0) = 0.6003345$, $E_0 = 1.59\times10^5$ Vm^{-1}; curve 2 corresponds to $(m_2/m_0) = 0.7812856$, $E_0 = 3.06\times10^5$ Vm^{-1} and curve 3 corresponds to $(m_2/m_0) = 2.1445069$, $E_d = 2.257\times10^5$ Vm^{-1}.

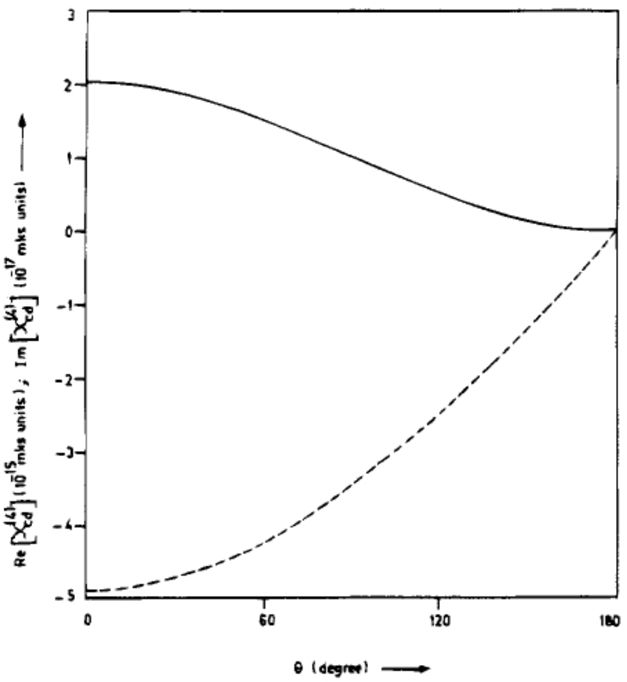

Figure 3: The variation of real (- - - -) and imaginary (-------) parts of $\chi_{cd}^{(4)}$ with the scattering angle θ for the following parameters: $E_d = 1.5\times10^3$ Vm^{-1}, for $E_0 = 1.59\times10^5$ Vm^{-1} and $(m_2/m_0) = 0.6003345$.

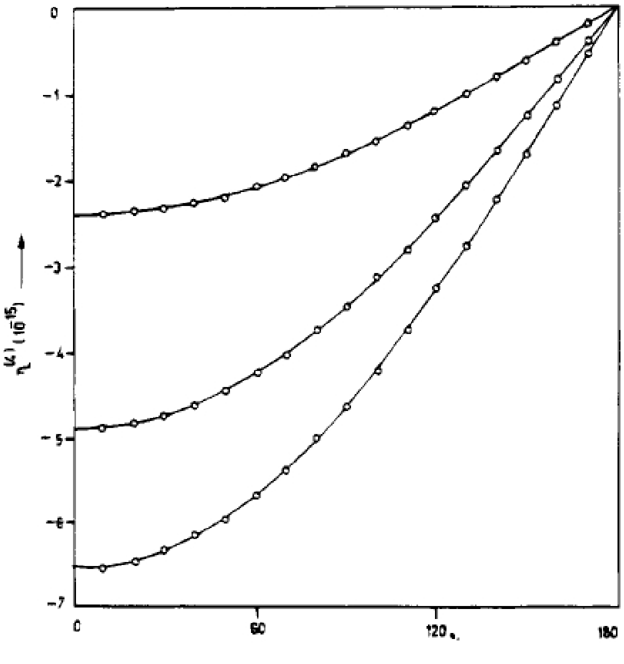

Figure 4: The variation of the nonlinear refractive index $\eta^{(4)}$ with the scattering angle θ for $E_d = 1.5\times10^3$ Vm^{-1}. The curve 1 corresponds to $(m_2/m_0) = 0.6003345$, $E_0 = 1.59\times10^5$ Vm^{-1}; curve 2 corresponds to $(m_2/m_0) = 0.7812856$, $E_0 = 3.06\times10^5$ Vm^{-1} and curve 3 corresponds to $(m_2/m_0) = 2.1445069$, $E_d = 2.257\times10^5$ Vm^{-1}.

Figure 2 shows the variation of the magnitude of the fourth order susceptibility as a function of scattering angle θ with (m_2/m_0) as a parameter. One can infer from Figure 2 that as the scattering angle increases, the magnitude of $\chi_{cd}^{(4)}$ decreases. From Figure 1 one finds that with the increase in the effective electric field (or amplitude of the pump wave) the value of (m_2/m_0) first decreases and after certain value it starts increasing. Similarly, by comparing all the curves of Figure 2 it can be inferred that with the increase in electric field amplitude, $\chi_{cd}^{(4)}$ first decreases and after a certain value it starts increasing, so by adjusting the value of E_0 or (m_2/m_0) one may achieve the desired value of nonlinear optical susceptibility in n-GaAs.

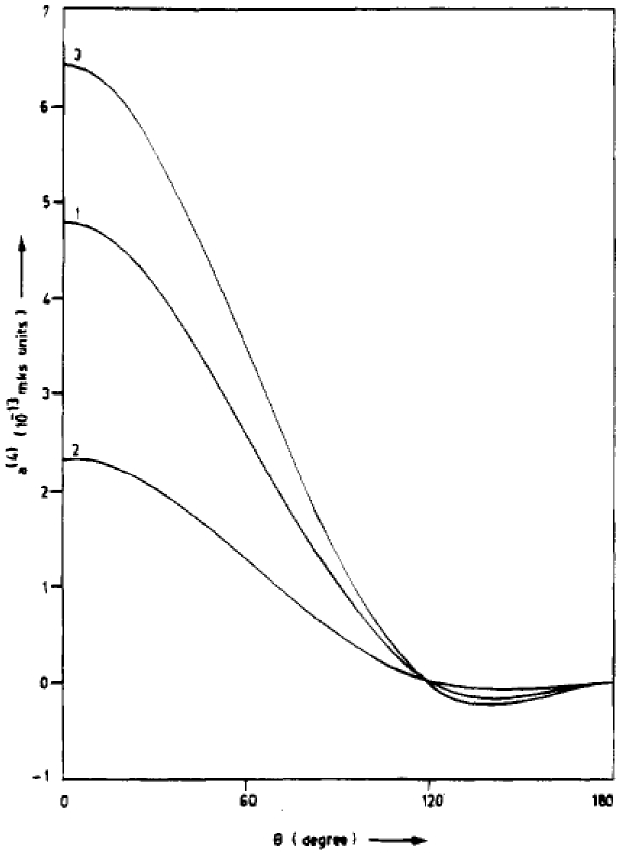

Figure 5: The variation of the nonlinear absorption coefficient $\alpha_{nl}^{(4)}$ with the scattering angle θ for $E_d = 1.5\times10^3$ Vm^{-1}. The curve 1 corresponds to $(m_2/m_0) = 0.6003345$, $E_0 = 1.59\times10^5$ Vm^{-1}; curve 2 corresponds to $(m_2/m_0) = 0.7812856$, $E_0 = 3.06\times10^5$ Vm^{-1} and curve 3 corresponds to $(m_2/m_0) = 2.1445069$, $E_d = 2.257\times10^5$ Vm^{-1}.

We have plotted the real and imaginary parts of fourth order susceptibility of the system as a function of scattering angles and the results are displayed in Fig. 3. It is found that as scattering angle increases, the real part of the susceptibility increases whereas the imaginary part of it decreases. We have displayed nonlinear refractive index and absorption coefficient as a function of θ using (m_2/m_0) as parameter in Figures 4 and 5, respectively. These figures reveal that with an increase in scattering angle, the refractive index increases while the absorption coefficient decreases. In other words with the increase in scattering angle the transmitted power increases. One may infer from Figure 5 that the absorption coefficient with any set of parameters becomes equal to zero at θ = 120° and then cross-over to a negative value for θ > 120°. It is a well-known fact that negative absorption coefficient yields gain. Thus one may adjust the value of the scattering angle θ between 120^o to 160^o to achieve a gain of the scattered mode. This type of behaviour of the absorption coefficient can be utilized in the construction of optical switches.

4. Conclusions

The above discussion reveals that a high power pump beam propagating through an n-GaAs sample under a d.c. bias decays into an acoustic wave and a scattered electromagnetic radiation. The nonlinearity arises through piezoelectricity and the field-dependent effective mass of electrons. It is found that the field dependency of effective mass is responsible for fourth order nonlinearity in the sample. Hence it is concluded that the negative differential resistivity region in n-GaAs has a great influence on the nonlinear optical parameters of the system.

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