Optical nonlinearity due to intersubband transitions in semiconductor quantum wells

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We have calculated the contribution of intersubband transitions to the third order optical nonlinear susceptibility, $\chi^{(3)}(\omega, \omega, \omega)$ for nonresonant as well as resonant third harmonic generation and $\chi^{(3)}(\omega, -\omega, \omega)$ for nonlinear refraction and absorption. As examples, we consider InAs/AlSb and GaAs/GaAlAs quantum wells. The effects of finite barrier height, energy band nonparabolicity, and high carrier concentrations are included. It is shown that quantum confinement, rather than the band nonparabolicity, is responsible for high values of nonresonant $\chi^{(3)}$. Very high values of $\chi^{(3)}$ are obtained for third harmonic generation and two photon absorption for incident wavelength near $10.6 \mu m$. Intensity dependence of refractive index and of absorption coefficient is also discussed for intensity well above the saturation intensity. Effective medium theory is used to incorporate the collective effects.

1. INTRODUCTION

Intersubband transitions (ISBT) in semiconductor quantum wells (QW) have attracted a lot of attention in nonlinear optics since the work of West and Eglash. As emphasized in a recent review by Almogy and Yariv, the main reason for this interest is the occurrence of narrow transitions with large oscillator strengths. In bulk semiconductors mobile carrier optical nonlinearity, $\chi^{(3)}$, due to intraband transitions is proportional to $\frac{1}{\hbar^2} \frac{\partial^2 E}{\partial k^2}$ at low frequencies, where photon energy $\hbar \omega \ll E_g$, the semiconductor bandgap; $E$ is the electron energy, and $k$ is the electron wave vector. This suggests that nonlinearity in bulk semiconductors increases with increase in the energy band nonparabolicity. Some attempts have been made to extend the above relation to calculate $\chi^{(3)}$ due to mobile carriers in a semiconductor QW with a view to enhance the nonparabolicity and hence the optical nonlinearity. However, as noted by Jha and Bloembergen, the low frequency limit is valid when $\hbar \omega$ is much smaller than the relevant bandgaps, which in the present case is the intersubband gap, $E_{ISBT}$. Thus in a superlattice (SL) or a multiple quantum well (MQW) there are two relevant low-frequency regimes: $E_{ISBT} \leq \hbar \omega < E_g$, and second, $\hbar \omega < E_{ISBT}$. For the former regime, which is of more practical interest, the above simple relation is not applicable and it is important to understand the role played by quantum confinement. Resonant enhancement of nonlinearity at ISBT is the second major theme in the exploitation of quantum wells as nonlinear materials. High values of $\chi^{(3)}$ have been reported by third harmonic generation (THG) involving ISBT resonances. The third important aspect of optical nonlinearities involving ISBT is the significant effects of electron-electron interaction. Two of the most important consequences of including the Coulomb interaction are the shift of the intersubband resonance due to depolarization effects and the predicted occurrence of optical bistability. Most of the work cited above has emphasized one or the other aspect. In this paper we develop a theory of the optical nonlinearities in the intersubband transition region including the essential complexities such as, finite barrier height which allows the electron wavefunctions to extend into the barrier region, nonparabolicity which affects the intersubband energies, the subband effective masses, and the collective effect which is important at high carrier concentrations. To do this we first calculate the single particle response including realistic subband dispersions and then include electron-electron interaction through an effective medium theory.

In Section II we present the theoretical formulation. Various approximations necessary to deal with nonparabolic bands in quantum wells are stated as well the necessary details of the expressions used for evaluating nonlinear susceptibilities. The nonlinear susceptibility $\chi^{(3)}$ due to intersubband resonance is denoted as $\chi^{(3)}(3\omega)$ hereafter denoted as $\chi^{(3)}(3\omega)$ by making use of third harmonic generation (THG) involving ISBT resonances. We show a very high value of $\chi^{(3)}(3\omega)$ (of the order of $10^{-4}$ e.s.u. at $10.6 \mu m$) can be achieved from this system. In Section IV, $\chi^{(3)}(-\omega)$, are calculated for the same material system and also for GaAs/GaAlAs systems. This material system has become important because of its high barrier height ($1.35$ eV) which allows large charge density (of the order of $10^{19}$ cm$^{-3}$) to accumulate. Very high room temperature mobility ($2 \times 10^4$ cm$^2$/V$^{-1}$s$^{-1}$), and ultra-fast energy relaxation time (1 ps at room temperature) of the system are also useful for device applications. We show a very high value of $\chi^{(3)}(3\omega)$ (of the order of $10^{-4}$ e.s.u. at $10.6 \mu m$) can be achieved from this system. In Section IV, $\chi^{(3)}(-\omega)$, are calculated for the same material system and also for GaAs/GaAlAs systems.
MQW system. We find that large value of $\chi^{(3)}(−\omega)$ can be achieved at two photon absorption (TPA) wavelength which is near $\sim 10.6\mu m$ for InAs/AlSb system. Since the values of $\text{Re}(\chi^{(3)}(−\omega))$ is larger than that of $\text{Im}(\chi^{(3)}(−\omega))$ near TPA wavelength useful applications of these systems in optical switching devices may be expected. Optical bistability has already been predicted for InAs/AlSb system after considering electron-electron interaction through the effective medium theory. This intrinsic optical bistability of a medium is of considerable theoretical interest.

In Section V the numerical results of intensity-dependent absorption including the power-broadening term are presented. The calculated values obtained by using the effective medium theory to include the depolarization effects compare very well with earlier experimental and theoretical results.

The conclusions are presented in Section VI.

II. THEORETICAL FORMULATION OF $\chi^{(3)}$

In this section we first formulate the single particle response $\chi^{(3)}$ for ISBT, and then the electron-electron interaction is included through the effective medium theory.

Single particle response

We consider a MQW system consisting of rectangular wells and barriers of widths $L_W$ and $L_B$, respectively. The well region is doped n-type so that at least the lowest subband is always populated. The z-direction is chosen to be perpendicular to the plane of the wells with $z = 0$ at the center of a well. The equation for the envelope function, $F(z)$ is then written as [4]

$$\frac{\hbar^2}{2m^*_E(E)} \frac{d^2 F_i(z)}{dz^2} + \left[ (E - E_{ci}) - \frac{\hbar^2 k_l^2}{2m^*_E(E)} \right] F_i(z) = 0,$$

where $m^*_E$ is the effective mass of the electron, $E$ is the energy eigenvalue including the component due to in-plane motion, and $k_l$ is the in-plane momentum of the electron; $m^*_E$ and $m^*_V$ are the energy and the velocity effective masses, respectively, and are given by

$$m^*_E(E) = m^*_e(0) \left\{ 1 + \alpha_i (E - E_{ci}) \right\}, \quad (3a)$$

and

$$m^*_V(E) = m^*_v(0) \left\{ 1 + 2\alpha_i (E - E_{ci}) \right\}, \quad (3b)$$

where $m^*_i(0)$ is the band-edge effective mass in the bulk material, and $\alpha_i$ is the nonparabolicity parameter of the $i$ layer.

We calculate the third order optical susceptibility, $\chi^{(3)}_{zzzz}$ in the z-direction. In this paper susceptibilities in the in-plane directions will not be considered. The expression for $\chi^{(3)}_{ijkl}(−\omega_1, \omega_2, \omega_3)$, where $\omega_p = \omega_1 + \omega_2 + \omega_3$ can be obtained by using the standard density matrix perturbation formalism [6] and has 48 terms which are reduced to 24 distinct terms for $i = j = k = l = z$.

Resonant third order susceptibilities have been calculated for the cases of THG and NRA by the sum-over-states method. For THG we get,

$$\chi^{(3)}_{zzzz} = \frac{e^4}{\pi \hbar^5 L_W} \times \int_0^\infty \sum_a m^*_i W(E) f(E) S(\omega) dE,$$

where $f(E)$ is the Fermi function given by

$$f(E) = \frac{1}{1 + exp\{(E - E_a - E_f)/k_B T\}}, \quad (4)$$

where $E$ is the energy corresponding to the in-plane momentum of the electron, $E_a$ is the energy eigenvalue of the state $a$ of the QW, and $E_f$ is the Fermi energy. The factor $S(\omega)$ in Eq. (4) is

$$S(\omega) = \sum_{b \neq a} \sum_{c \neq a} \sum_{d \neq a} \sum_{e \neq a} z_{ab} z_{bc} z_{cd} z_{da}$$

$$\times \left\{ \left( \frac{\omega_{ba} - 3\omega \omega_{ca} - 2\omega \omega_{da} - \omega}{\omega_{ba} + \omega} \right) \left( \frac{\omega_{ca} + 2\omega \omega_{da} - \omega}{\omega_{ca} + \omega} \right) \right\}$$

$$- \left\{ \sum_{b \neq a} \sum_{d \neq a} \left| z_{ab} \right|^2 \left| z_{ad} \right|^2$$

$$\times \left\{ \left( \frac{\omega_{ba} - 3\omega \omega_{da} - \omega}{\omega_{ba} + \omega} \right) \left( \frac{\omega_{da} - 3\omega}{\omega_{da} + 3\omega} \right) \right\} \right\}$$

where $z_{jk}$ are dipole matrix elements between two envelope states in the $j$-th and $k$-th subbands. In Eq. (4), $\hbar \omega_{jk} = E_{jk} - i \Gamma_{jk}$, where $E_{jk}$ is the intersubband energy
between the states \( j \) and \( k \), and \( \Gamma_{jk} \) is the broadening parameter; and \( \omega_{jk}^* \) is the complex conjugate of \( \omega_{jk} \). In general, \( z_{jk} \) and \( E_{jk} \) depend on the in-plane momentum of the electron for nonparabolic bands.

The NRA are the other important processes which has not been discussed in detail in the literature.\(^{24}\)

The importance of a complete theoretical expression of \( \chi_{zzzz}^{(3)}(\omega, \omega, -\omega, \omega) \) was pointed out earlier by Bloembergen et. al.\(^{23}\) for a system where close one and two photon resonances were present. In this article we present a complete expression of \( \chi_{zzzz}^{(3)}(\omega, \omega, -\omega, \omega) \) due to ISBT for a heavily doped MQW system in which resonances are similar to the case considered by Bloembergen et. al.\(^{23}\)

The third order susceptibility for NRA is given by

\[
\chi_{zzzz}^{(3)}(\omega, \omega, -\omega, \omega) = \frac{e^4}{3\pi\hbar^3 L_W} \times \int_0^\infty \sum_a m_{VW}^a(E)f(E)T(\omega)dE, \quad (7)
\]

where

\[
T(\omega) = \sum_{b \neq c} \sum_{d \neq a} z_{ab}z_{bc}z_{cd}z_{da} \times \frac{1}{(\omega_{da} - \omega)\omega_{ca}(\omega_{ba} - \omega)} + \frac{1}{(\omega_{da} - \omega)\omega_{ca}(\omega_{ba} + \omega)} + \frac{1}{(\omega_{da} - \omega)(\omega_{ca} - 2\omega)(\omega_{ba} - \omega)} + \frac{1}{(\omega_{da} - \omega)(\omega_{ca} - 2\omega)(\omega_{ba} + \omega)} + \frac{1}{(\omega_{da} + \omega)(\omega_{ca} - 2\omega)(\omega_{ba} - \omega)} + \frac{1}{(\omega_{da} + \omega)(\omega_{ca} - 2\omega)(\omega_{ba} + \omega)} + \frac{1}{(\omega_{da} - \omega)(\omega_{ca} + 2\omega)(\omega_{ba} - \omega)} + \frac{1}{(\omega_{da} + \omega)(\omega_{ca} + 2\omega)(\omega_{ba} + \omega)} - \sum_b \sum_d |z_{ab}|^2 |z_{ad}|^2 \times \left\{ \frac{1}{(\omega_{da} - \omega)(\omega_{ba} - \omega)(\omega_{ba} - \omega)} \right\} \quad (8)
\]

The terms following the double summations in the expressions for \( S(\omega) \) and \( T(\omega) \) are the separated out terms containing the ground state as an intermediate state \((c = a)\). The apparently divergent terms \((c = a)\) in the expression for \( T(\omega) \) are reduced in the usual manner.\(^{23}\)

It should be noted that the expression for \( \chi_{zzzz}^{(3)}(-\omega) \) is complete and is rigorously calculated from the original 24 terms which are obtained from the perturbation theory in dissipative systems. These 24 terms then reduce to 12 terms due to \( \omega_1 = -\omega_2 = \omega_3 = \omega \). Since the imaginary part of \( \chi_{zzzz}^{(3)}(-\omega) \) gives the two photon absorption (TPA) co-efficients, it should be positive for the entire frequency range. Taking the proper sign of the damping factor is, therefore, important. In the calculation of THG, the terms are calculated with the help of the perturbation theory in the absence of damping, and then the damping factors are introduced at the final expression to obtain the Lorentzian broadening at \( \omega \), \( 2\omega \), and \( 3\omega \) resonances.\(^{24}\)

It was shown\(^{23}\) that in the THG dispersion, the term containing \((|\omega_{ba} - 3\omega|^2)/(\omega_{ca} - 2\omega)(\omega_{da} - \omega)^{-1}\) is dominant among all the 48 terms and no cancellation of resonant terms is involved. Hence our approximation for \( \Gamma_{ij} \) is justified.

The nonresonant optical nonlinearity can be calculated from Eq. (3) by putting \( \Gamma = 0 \) taking the limit, \( \omega \to 0 \). The resultant expression is

\[
\chi_{zzzz}^{(3)}(0) = \frac{4e^4}{h^3\pi L_W} \times \int_0^\infty \sum_a m_{VW}^a(E)f(E)
\]
\[
\chi^{(3)}_{zzzz}(0) = \frac{e^4 N m^3 W}{32 \hbar^6} \frac{L^9_W}{N} \sum_{n=1}^{N} \left( -\frac{2}{9 \pi^6 n^6} + \frac{140}{3 \pi^8 n^8} - \frac{440}{\pi^{10} n^{10}} \right)
\]
where \(N_s\) is the sheet carrier concentration, \(m^*_W\) is the bulk band edge effective mass and \(N\) is the total number of occupied subbands.

To calculate the intensity-dependent absorption we have included power broadening following the general formalism\(^\text{[2]}\) to obtain the expression for the total (linear and nonlinear combined) optical susceptibility, \(\chi(-\omega)\) as:

\[
\chi(-\omega) = -\frac{e^2}{\epsilon_0 L_W \pi \hbar^2} \int_0^\infty \sum_a \sum_b \left( m^*_W(E)f_a(E) - m^*_W(E)f_b(E) \right) \frac{\varepsilon_{ab}z_{ba}(\hbar \omega - E_{ba} - i\Gamma_2) \Gamma_2^{-2}}{1 + (\hbar \omega - E_{ba})^2 \Gamma_2^{-2} + 4\varepsilon_{ab}z_{ba}|E|^2 \Gamma_1 \Gamma_2^{-2}} \, dE
\]

where \(a\) and \(b\) are two consecutive energy levels \((b > a)\), \(\Gamma_1 = \hbar / T_1\), and \(\Gamma_2 = \hbar / T_2\) with \(T_1, T_2\) being the lifetime and the dephasing time, respectively.

**Electron-electron interaction**

It is generally accepted that the Coulomb interaction between the carriers plays an important role in the enhancement of optical nonlinearity and the blue shift of intersubband resonances. For many systems, e.g. a metallic sphere, or a metal-insulator interface simple electromagnetic theories of surface plasmons and surface polaritons have been important in understanding this interaction\(^\text{[4]}\). The effective medium theory is a similar one which we will apply for MQWs in this section. The direct physical nature of this theory allows us to predict the plasmon frequency-dependence on physical parameters, such as the width and the dielectric constant of the barrier material. For low carrier concentration the modification from single electron approximation is not so important, on the other hand, for high carrier concentrations collective effects are important\(^\text{[4]}\). We have already shown that the effective medium theory can give a good representation of the collective effects\(^\text{[1]}\) in linear absorption spectra of ISBT\(^\text{[1]}\). Here we extend the effective medium theory to nonlinear processes and show good agreements between our calculated results and experimental observations\(^\text{[1]}\) as well as theoretical results\(^\text{[4]}\) reported earlier. These experimental results are otherwise explainable only when many-electron calculations are done\(^\text{[4]}\). This is an indication that the effective medium theory is capable of representing the most important many-electron effects in such systems in a direct and physically attractive way. Recent calculations by Das Sarma and Hwang\(^\text{[6]}\) also show the importance of the barrier width in understanding the behavior of plasmons in MQWs, which is in agreement with our results.

We take a MQW system as a layered medium with alternate layers of width \(L_W = ad\) and \(L_B = (1-\alpha)d\) where \(d\) is the period of the MQW and \(\alpha\) is a fraction. For \(d \ll \lambda\), the wavelength of light, the composite medium can be described by an effective dielectric function, \(\tilde{\epsilon}_{ij}(\omega)\). The calculation of the nonlinear response has to be performed in a self-consistent manner since the electric field in the well region, \(E_W\) and in the barrier region, \(E_B\) depend on the dielectric functions as

\[
E_W = \epsilon_B \tilde{E}/\{\epsilon_B \alpha + \epsilon_W (1-\alpha)\},
\]
and

\[
E_B = \epsilon_W \tilde{E}/\{\epsilon_B \alpha + \epsilon_W (1-\alpha)\},
\]

where \(\tilde{E}\) corresponds to the average electric field in the \(z\)-direction given by

\[
\tilde{E} = \alpha E_W + (1-\alpha) E_B.
\]

The function \(\epsilon_W\), the dielectric permittivity of the well layer, in turn, depends on \(E_W\) according to \(\epsilon_W = \epsilon_{bulk} + \epsilon_{intersubband}\) where \(\epsilon_{intersubband}\) is calculated from Eq. \(\text{[1]}\). For simplicity we have assumed that \(\epsilon_B\), the dielectric permittivity of the barrier layer is a constant in the frequency range of interest. In case of \(z\)-polarized light, the electric displacement vector across the interface is continuous, i.e.

\[
\epsilon_W E_W = \epsilon_B E_B = \tilde{\epsilon} \tilde{E}.
\]

From Eqs. \(\text{[1]}\), \(\text{[3]}\), and \(\text{[4]}\) we obtain the average dielectric constant of the composite medium as

\[
\tilde{\epsilon} = \frac{\epsilon_W \epsilon_B}{\alpha \epsilon_B + (1-\alpha) \epsilon_W}.
\]

From Eqs. \(\text{[1]}\), \(\text{[4]}\), and \(\text{[5]}\) we obtain the following relation:

\[
P^2 C^2 x^3 + \left[ 2 R e (PC \beta_0 e^{i\phi}) + 2 P^2 CB - y C^2 \right] x^2 + \left[ B^2 P^2 + \beta_0^2 + 2 R e (BP \beta_0 e^{i\phi}) \right] x - y B^2 = 0,
\]

where
Using Eq. (10)

\[ P = \frac{\alpha}{1 - \alpha} \epsilon_B + \epsilon_{bulk} + 1, \]
\[ B = 1 + (\hbar \omega - E_{ba})^2 \Gamma_1 \Gamma_2, \]
\[ C = 4 z_{ab} z_{ba} (\Gamma_1 \Gamma_2)^{-1}, \]
\[ \beta_{0,16} = -\frac{e^2}{\epsilon_0 L_W} |n_{sa} - n_{sh}| |z_{ba}|^2 (\hbar \omega - E_{ba} - i \Gamma_2) \Gamma_2^{-2}, \]
\[ y = \left| \frac{\epsilon_B}{1 - \alpha} \right|^2, \]
\[ x = |E_W|^2. \]

In the above \( n_{si} \) is the sheet carrier concentraion of the \( i \)-th energy level. Equation (16) admits multivalued solutions for intensity in the well region as a function of \( i \). In the above saturation intensity can be calculated from

\[ \chi(-\omega) = \chi(1)_{zz}(-\omega) + 3\chi(3)_{zzzz}(\omega) |E_W|^2, \]

where \( \chi(1)_{zz}(-\omega) \) is the linear susceptibility due to ISBT. In this case, the collective effect can be introduced following the formulation as reported elsewhere.\[1\]

III. NONRESONANT \( \chi^{(3)}(0) \)

The nonresonant third order nonlinearity \( \chi^{(3)}_{zzzz}(0) \) due to ISBT has been calculated using Eq. (16) for an InAs/AlSb MQW system used by Warburton et al.\[2\]. The well and the barrier widths are 18 and 10 nm, respectively, and the carrier concentration is 2.61 \( \times \) 10\(^{12} \) cm\(^{-2} \) at 10K whereby the second subband is populated. The values of the physical parameters for the calculations, and the results are given in Tables I and II, respectively.

| Material Systems | \( m_W/m_0 \) | \( m_B/m_0 \) | \( \omega_w \) | \( \alpha_B \) | \( E_{CB} - E_{CW} \) |
|------------------|----------------|----------------|-----------|--------|----------------|
|                   | (eV\(^{-1} \)) | (eV\(^{-1} \)) | (eV)      |        | (eV)          |
| InAs/AlSb        | 0.025          | 0.260          | 2.050     | 0.223  | 1.350          |
| GaAs/GaAlAs      | 0.067          | 0.0919         | 0.573     | 0.295  | 0.261          |

\( m_0 \) is the free electron mass.

| Table II. Calculated values of \( \chi^{(3)}(0) \) in e.s.u. |
|-----------------|----------------|----------------|
| Parabolic       | Nonparabolic   | Using Eq. (16) |
| \( -9.80 \times 10^{-1} \) | \( -5.08 \times 10^{-1} \) | \( -5.27 \times 10^{-3} \) |

The values of \( \chi^{(3)}_{zzzz}(0) \) are negative in all cases whereas the low-frequency mobile carrier contribution to \( \chi^{(3)} \) for bulk InAs is positive. It is found that \( \chi^{(3)}_{zzzz}(0) \) is non-zero even for the parabolic band approximation, which suggests that nonlinearity is mainly due to confinement of electron wave function within the quantum well region. When nonparabolicity is included, the dipole matrix elements do not change much,\[1\] but the zone centre ISBT energies decrease compared to those for the parabolic band approximation, and continue to decrease with increase in the in-plane momentum. As a result, both the terms within the parenthesis in Eq. (16) increase. But, the rate of increase of the first term which is smaller of the two terms, is more than that of the second. Hence \( |\chi^{(3)}_{zzzz}(0)| \) decreases.

It is instructive to compare these results with the values of \( \chi^{(3)}_{zzzz}(0) \) calculated for infinitely deep QWs using Eq. (16). We see that the value of \( |\chi^{(3)}_{zzzz}(0)| \) is much greater than that obtained by using Eq. (16) for infinite barrier model. This is expected for two reasons. Firstly, since a real-life QW system has a finite height, the electron wavefunction penetrates into the barrier region. As the electron spends part of the time in the barrier layer, the effective well width increases, and \( |\chi^{(3)}_{zzzz}(0)| \) being proportional to the ninth power of the well width, for finite barrier heights, its value increases by an order of magnitude even for the parabolic band approximation. Secondly, in the calculation using Eq. (16) partially occupied subbands are not included, i.e. only the transitions at or near the zone centre are considered. This precludes transitions from the first to the second subband when the latter is populated. However, such transitions do occur\[4\] for \( k_{f2} < k_\perp < k_{f3} \) where \( k_{f1} \) and \( k_{f2} \) are the Fermi wave vectors for the first two subbands. Such transitions have been included in Eq. (16), which leads to an increase in the value of \( |\chi^{(3)}_{zzzz}(0)| \).

From these results we may conclude that nonlinearity due to ISBT is influenced by both the band nonparabolicity as in bulk semiconductors, and the quantum confinement of the electron wavefunction. The main contribution is from the quantum confinement which is substantially modified by nonparabolicity.

IV. THIRD HARMONIC GENERATION, \( \chi^{(3)}_{zzzz}(3\omega) \)

The real and the imaginary parts of \( \chi^{(3)}_{zzzz} \) due to THG are shown in Fig 1. The calculation have been done for InAs/AlSb MQW system by sum-over -states method. There are 14 bound levels, all the levels are taken into account in calculations. It is seen that the value \( \chi^{(3)}(3\omega) \) converges after taking 10 bound levels. Because sum of the oscillator strengths for the bound states becomes close to unity, the effect of continuum may be safely neglected. In Fig 2. \( |\chi^{(3)}_{zzzz}(3\omega)| \) is shown, which is a measurable quantity. There are 8 peaks due to \( \omega, 2\omega, \) and \( 3\omega \) res-
onances from the two occupied subbands. The $\omega$, $2\omega$, and $3\omega$ resonances due to transitions from the ground subband occur at $h\omega = 77, 95, 25$, and $105$ meV, respectively. The resonances due to transitions from the first excited level occur at $h\omega = 110, 117, 36$, and $120$ meV, respectively. The peaks at $25$ and $36$ meV are very small, as shown in the figure. It is seen that around $117$ meV, i.e., corresponding to the $10.6$ $\mu$m region, a very high value, about $4.4 \times 10^{-4}$ e.s.u. of $|\chi_{zzzz}(3\omega)|$ is obtained, which is the largest value of nonlinearity due to THG ever reported for a single symmetric QW system. Since this MQW system already exists, it is worthwhile to do experiments to establish high nonlinearities of purely electronic origin. We note that when depolarization effects are included, one-photon resonances would shift from the calculated positions as in one photon spectra.

V. NONLINEAR REFRACTION AND ABSORPTION, $\chi_{zzzz}^{(3)}(-\omega)$

The real and the imaginary parts of $\chi_{zzzz}^{(3)}(-\omega)$ are shown in Fig. 3. The imaginary part describes TPA. Here we obtain four major peaks due to transition from the ground and the first excited state.

![FIG. 3. Dependence of $\chi_{zzzz}^{(3)}(-\omega)$ on the incident wavelength for the InAs/AlSb MQW system. The dotted and the solid lines show the real and the imaginary parts, respectively.](image)

From the figure it is seen that at about $10.6$ $\mu$m there is a resonance due to two photon absorption between the second and the fourth subbands. The magnitude of third order optical nonlinearity corresponding to this TPA is also very high, of about $4 \times 10^{-4}$ e.s.u. It is comparable to the largest reported nonlinearity at $10.6$ $\mu$m for $\text{Hg}_{0.84}\text{Cd}_{0.16}\text{Te}$ (Ref. 32). It should be noted that this resonant nonlinearity is of practical interest because no linear absorption occurs at this wavelength. For all optical switching devices the ratio, $\left|\frac{\text{Re}(\chi_{zzzz}^{(3)})}{\text{Im}(\chi_{zzzz}^{(3)})}\right|$ representing the phase of the nonlinearity is important, and should have a value greater than 2 (Ref. 15,18) for device applications. This ratio is plotted in Fig. 4, and we see that the condition is satisfied near the TPA wavelength. We have earlier predicted\cite{4} intrinsic optical bistability in this sample considering electron-electron interaction through effective medium theory.

The real and the imaginary parts of $\chi_{zzzz}^{(3)}(-\omega)$ of GaAs/GaAlAs MQW system are plotted in Fig. 5. The physical parameters used in the calculations correspond to the sample used by Craig et al.\cite{2} are listed in Table I.

![FIG. 2. Dependence of $|\chi_{zzzz}^{(3)}(3\omega)|$ on the photon energy for the InAs/AlSb MQW system.](image)

The results for the higher carrier concentration ($N_s = 1.1 \times 10^{11}$ cm$^{-2}$) are given in this figure. Since only one energy level is populated here, two peaks, one each for one photon and two photon resonances are observed. The high value of $\chi_{zzzz}^{(3)}(-\omega)$ ($2.2 \times 10^{-2}$ e.s.u.) at the TPA frequency is due to the large well width of 40 nm and
high carrier concentration. Since \( \left| \frac{\text{Re}(\chi^{(3)})}{\text{Im}(\chi^{(3)})} \right| > 2 \) (see Fig. 6) near the TPA frequency, this particular QW system is also expected to be useful for optical switching.

![Graph](image1.png)

**FIG. 4.** Dependence of \( \left| \frac{\text{Re}(\chi^{(3)}(-\omega))}{\text{Im}(\chi^{(3)}(-\omega))} \right| \) on the incident wavelength for the InAs/AlSb MQW system.

![Graph](image2.png)

**FIG. 5.** Dependence of \( \chi^{(3)}(-\omega) \) on the photon energy for the GaAs/GaAlAs MQW system. The dotted and the solid lines show the real and the imaginary parts, respectively.

Using Eq. (7) we have also calculated \( |\chi^{(3)}_zzzz(-\omega)| \) for GaAs/GaAlAs MQW systems used by Walrod et al. and obtained a set of values of \( 3.65 \times 10^{-5} \) and \( 3.91 \times 10^{-5} \) e.s.u. which are in good agreement with their experimental results, \( 3.3 \times 10^{-5} \) and \( 4.25 \times 10^{-5} \) e.s.u., respectively.

![Graph](image3.png)

**FIG. 6.** Dependence of \( \left| \frac{\text{Re}(\chi^{(3)}(-\omega))}{\text{Im}(\chi^{(3)}(-\omega))} \right| \) on the photon energy for the GaAs/GaAlAs MQW system.

### VI. INTENSITY-DEPENDENT ABSORPTION

To calculate the intensity-dependent absorption we consider the GaAs/AlGaAs system used by Craig et al. The values of transition electric dipole moments and the dephasing relaxation time, \( T_2 \) are taken from the experimental work. We also assume \( T_1 = T_2 \). In the effective medium theory the intensity in, and the intensity-dependent dielectric function of the well region depend on each other through Eqs. (11) and (12). These were solved self-consistently at each intensity to obtain \( \tilde{\epsilon} \) whose imaginary part determines the absorption by the composite medium. These results are shown in Fig. 7.

![Graph](image4.png)

**FIG. 7.** Dependence of scaled absorption on the frequency of the incident radiation for the GaAs/GaAlAs MQW system for \( T_1 = T_2 \). The intensities of the radiation for the highest to the lowest peaks are \( 10^{-3}, 100, 200, 500, 800, \) and \( 1000 \) Wcm\(^{-2} \), respectively.

Here, the absorption co-efficient, multiplied by a scale factor, \( \hbar n_0 c/q \) where \( n_0 \), \( c \), and \( q \) are the refractive index
of bulk GaAs, velocity of light, and the electronic charge, respectively is plotted in the ordinate. The asymmetry in the lineshapes of the absorption curves is known to occur due to the collective effect and is well reproduced in our calculation. We note that the observed position of the linear absorption peak at 81 cm$^{-1}$ is blue shifted from the calculated one electron peak position at 72 cm$^{-1}$, which can be explained as the depolarization shift with the help of the many electron theory. This shift has also been accurately reproduced in our calculations. Using an intensity-dependent $T_1$ it is possible to get much better agreement with the experimental intensity-dependent absorption spectrum for the sample having $N_s = 1.1 \times 10^{11}$ cm$^{-2}$. For the other samples, a change in the absorption frequency by 5 cm$^{-1}$ is observed for high incident intensities. The discrepancy is due to the asymmetry in the QW shape caused by an applied DC electric field which has not been included in our calculations. We have also obtained similar results for the GaAs/GaAlAs system discussed by Zaluzny.

VII. CONCLUSIONS

In this paper we have presented detailed calculations of the third order optical nonlinearity due to ISBT in semiconductor MQW systems. We first calculate the single particle linear and nonlinear response functions. The most important effects of electron-electron interactions i.e. the screening due to depolarization terms are included then through an effective medium theory. In calculating the single particle response all the essential complexities, such as the finite barrier height and the energy band nonparabolicity are included. Very high values of $\chi^{(3)}_{zzzz}(-\omega)$ ($4 \times 10^{-4}$ e.s.u.) and $\chi^{(3)}_{zzzz}(3\omega)$ ($4.4 \times 10^{-4}$ e.s.u.) are predicted near 10.6 $\mu$m for the InAs/AlSb MQW system. We find that in the low-frequency regime the nonlinearity is mainly due to quantum confinement of electrons - the effect of nonparabolicity being a modification of this nonlinearity.

The inclusion of electron-electron screening terms through an effective medium theory gives rise to several new features. In linear spectra the dominant absorption peaks are attributed to intersubband plasmons. The blue and the red shifts observed in the linear and the nonlinear optical absorptions, respectively due to the many electron effect can be explained in terms of the effective medium theory. The asymmetry of the lineshapes of the nonlinear absorption spectra for intensities higher than the saturation intensity is also obtained.

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