Intensity-dependent nonlinear optical properties in an asymmetric Gaussian potential quantum well-modulated by external fields

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Abstract
In this paper, the effects of external electric, magnetic and non-resonant intense laser fields on the nonlinear optical rectification (NOR), second-harmonic (SH), and third harmonic (TH) generation in a GaAs quantum well with asymmetrical Gaussian potential are theoretically investigated. Firstly, the energy eigenvalues and eigenfunctions of a single electron confined in the structure are obtained by using the diagonalization method within the framework of the effective-mass and parabolic band approaches. Then, using these energy eigenvalues and eigenfunctions, expressions derived within the compact density matrix approximation have been employed to calculate the coefficients of the nonlinear optical response in the structure. The obtained simulation results show that the influence of the external fields leads to significant changes in the coefficients of nonlinear optical rectification, second and third harmonic generation in the system. As a result, it has been seen that the amplitude and position of the peaks of nonlinear optical rectification, second and third harmonic coefficients can be controlled by changing the applied external fields.

Keywords Asymmetric gaussian potential · Intense laser field · Second-harmonic generation · Third-harmonic generation · Nonlinear optical properties

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1 Introduction

The physical properties of nanomaterials have made them promising candidates for potential applications in modern device technology. For years, many researchers have investigated the optical and electronic characteristics of these materials. Among these works, one may find studies on the quantum plasmon effect (Scholl et al. 2012; Tan et al. 2014), the nonlinear optical effect (Guo and Gu 1993; Wang 2005; Zhang and Xie 2003), and magnetoresistance effects (Murzina et al. 2001). The most important feature that distinguishes nanomaterials from bulk materials is that the existence of unique optical and electronic properties, related to quantum confinement effects. Recently, remarkable progress has been made in artificial material growth technology. Thanks to this progress, the production process of nanomaterials can be achieved with nanometer-scale precision and resolution. Besides, it is now possible to prepare a variety of structures that distinguish each other by means of structural, namely in size and shape, and compositional parameters. The control of these parameters allows the analysis of optical and electronic properties in the nanomaterials.

The low dimensional semiconductor nanostructures are promising candidates for designing and fabricating electronic and optoelectronic devices. These structures are quantum wells (QWs), quantum well wires (QWWs), quantum dots (QDs), and also quantum rings (QRs). Among these structures, QWs having distinct confining potential shapes (i.e. square, parabolic, graded, and so on) can be experimentally obtained by using modern crystal growth procedures such as the molecular beam epitaxy (MBE) techniques as well as metal–organic chemical vapor deposition (MOCVD), and metal–organic vapor phase epitaxy (MOVPE). There have been several experimental studies that the different potential profiles have been used. In the experimental studies, Palmgren et al. investigated optical transitions of GaAs V-groove quantum wires using the MOVPE system (Palmgren et al. 2006). The effects of the applied external fields on the CdSe nanowires have been studied by Protasenko et al., and the optical emission of this structure has been analyzed (Protasenko et al. 2007). The growth of GaAs and GaP nanowires by MBE have been studied by photoluminescence (PL) and PL excitation (PLE) (Weman et al. 2009). Karlsson et al. explored the exciton transfer between two parallel quantum wires (Karlsson et al. 2007). To understand the physical mechanism of the experimental process, theoretical studies play a critical role to provide a detailed description of the physical origin of the process. Therefore, several theoretical investigations have been conducted on the above-mentioned nano systems (Huang 2008; Liu et al. 2013; Rezaei et al. 2010; Duque et al. 2010, 2013; Rezaei and Shojaeian Kish 2013; Yuan et al. 2013). Feddi et al. studied the influence of magnetic field in quantum crystals (Feddi et al. 2000) and the polarizability of bound polarons (Feddi et al. 2003). Dujardin et al. explored the electric field effect in a thin quantum disk (Dujardin et al. 2012). The effect of electric field on the energy of an off-center door in the quantum crystal has been explored by Assaid et al. (2001). Linear and nonlinear optical properties (Haouari et al. 2017) and linear and nonlinear magneto-optical properties of a spherical core/shell quantum dots (Feddi et al. 2017) have been explored.

Also, the nonlinear optical properties of these semiconductors have the potential for high technological device applications, namely infrared laser (Hakki and Paoli 1975), infrared detectors (Likharev 1998), and optical modulators (Leobandung et al. 1995). To understand the electronic and optical properties of nanomaterials, it is necessary to determine their behavior in presence of external fields. As it is known, the nonlinear optical effect is very small in symmetrical structures (Khurgin 1988). Thus, to generate a nonlinear optical response,
the symmetry of the structure is required to be broken by either material growth technology or applying an external field (Zhang and Xie 2003). For that reason, many researchers have investigated the nonlinear optical properties of non-symmetric nanostructures as are the cases of semi-parabolic quantum wells (Zhang and Xie 2003), parabolic quantum dots (Li et al. 2007; Şakiroğlu et al. 2012), semi-exponential quantum wells (Chen et al. 2008). These studies revealed that controlling the electric field and adjusting the structure parameters result in an efficient second-harmonic generation. The actual confinement potentials in the conduction and valence bands of GaAs/AlGaAs and InGaAs/GaAs/AlAs heterostructures often resemble the parabolic potential (Maksym and Chakraborty 1990; Drexler et al. 1994; Ashoori et al. 1992). However, the experimental studies have shown that the confinement potential is close to the Gaussian potential (Heitmann et al. 1997). So, in this work, a simulation is performed to examine the effect of external fields on the nonlinear optical properties of a GaAs QW with asymmetric Gaussian confinement potential. The simulation parameters are the intensities of the external electric field, the magnetic field, and the intense laser field (ILF), respectively. The effects of these external probes on the NOR, SHG, and THG coefficients of the system have been examined in detail in this study. For this reason, firstly, the Schrödinger equation is solved numerically to find the wave functions and the energy eigenvalues of the confined electrons in the conduction band. Then, the nonlinear optical properties of the system are calculated using this information about the electronic states. This paper is organized as follows: In Sect. 2 the theoretical equations we use to obtain the electronic and optical properties of the structure are given in detail. In Sect. 3 presents numerical results and discussions. Finally, a summary of the physical results obtained in the study is given in chapter 4.

2 Theory

In the theoretical model, we consider a GaAs QW with asymmetrical Gaussian potential (AGPQW) exposed to the above-mentioned electromagnetic probes. We assume that the structure extends along the z-axis. In the theoretical calculation, the non-perturbative theory describing atomic behavior is used to account for the effect of the high-frequency ILF on the structure (Sakiroglu et al. 2012; Ungan et al. 2010; Eseanu 2010). In the framework of the effective mass and parabolic band approximations, the Hamiltonian operator for an electron confined in the AGPQW in the presence of a magnetic field perpendicular to the growth direction \((B = (B, 0, 0))\), z-oriented static electric field \((F = (0, 0, F))\), and linearly polarized ILF parallel to the growth direction is given by

\[
H = -\frac{1}{2m^*}(P + eA(r))^2 + V_{AGPQW}(z, a_0) + eFz, \tag{1}
\]

where \(m^*, P, e, \text{ and } c\) are the effective mass of the electron, electron momentum operator, electron charge and speed of light, respectively. \(A(r) = (0, -Bz, 0)\) is the magnetic vector potential \((B = \nabla \times A)\), which specifies the electric field, and \(F\) is the strength of the electric field. The laser-dressed confinement potential is given by

\[
V_{AGPQW}(z, a_0, p) = \frac{eF_0}{m^*\sigma} \frac{2\pi}{\sigma} \int_0^{2\pi/\sigma} V(z + a_0 \sin \sigma t) dt, \tag{2}
\]

where \(a_0 = \frac{eF_0}{m^*\sigma^2}\) is the laser dressing parameter, which is proportional to the field strength \(F_0\), with non-resonant frequency \(\sigma\). That is, it is assumed that values of \(\sigma\) lie well above the typical intersubband frequencies in the system. In Eq. 2, the period of the radiation
\( \left( \frac{2\pi}{\omega} \right) \) is such that the system only feels the time average of the oscillating field. The confinement potential in the absence of any field is given by

\[
V_{AGPQW}(z) = \begin{cases} 
0 & z < 0 \\
-V_0 e^{-z^2/2L^2} & z \geq 0 
\end{cases}
\]

where \( V_0 \) is the confinement potential height, and \( L \) is the confinement potential width.

The optical properties of the structure can be obtained after solving for the allowed quantum states in AGPQW. The eigenproblem associated with the energy operator of the system (Eq. 1) is tackled by using the diagonalization method described in Ref. (Mora-Ramos et al. 2013). This approach implies the Fourier expansion of the state vector over a basis of orthonormal functions with the subsequent diagonalization of the resulting Hamiltonian matrix. This process allows obtaining the energy levels and corresponding wave functions. Then, the optical rectification, second-harmonic generation, and third-harmonic generation coefficients are derived by using the compact density matrix method and iterative procedure, to identify the relation of the dielectric response of the system associated to the subband spectrum - excited by the field of an external electromagnetic wave of frequency \( \omega \) and polarization along the growth direction. The electric field vector of the applied electromagnetic field can be written as

\[
E(t) = E_0 \cos (\omega t) = \tilde{E}^r e^{-i\omega t} + \tilde{E}^i e^{i\omega t},
\]

Then, the electronic polarization of the system under the field is expressed by Mou et al. (2014); Zhai (2014).

\[
P(t) = \varepsilon_0 \chi^{(1)}(\omega) \tilde{E} e^{i\omega t} + \varepsilon_0 \chi^{(2)}(\omega) \tilde{E}^2 e^{i2\omega t} + \varepsilon_0 \chi^{(3)}(\omega) \tilde{E}^3 e^{i3\omega t} + \ldots
\]

where \( \varepsilon_0 \) is vacuum permittivity. \( \chi^{(1)}(\omega), \chi^{(2)}(\omega), \chi^{(3)}(\omega) \) and \( \chi^{(3)}(\omega) \) are the different contributions to the dielectric susceptibility, corresponding to linear, optical rectification, second harmonic generation, third-order, and third harmonic generation terms, respectively. The expression for the NOR, SHG, and THG susceptibilities are given below (Wang 2005; Martínez-Orozco et al. 2012; Mora-Ramos et al. 2012; Yu and Wang 2011)

\[
\chi^{(2)}(\omega) = \frac{4\varepsilon^2 \sigma_v^2 M^2_{01} \delta_{01} \omega_{10}^2 (1 + \Gamma_2/\Gamma_1) + (\omega^2 + \Gamma_2^2) \Gamma_2 / \Gamma_1 - 1}{(\omega_{10} - \omega)^2 + \Gamma_3^2 \Gamma_2 \Gamma_3}.
\]

\[
\chi^{(2)}_{2\omega} = \frac{\varepsilon^2 \sigma_v M_{01} M_{12} M_{20}}{\varepsilon_0 \hbar (\omega - \omega_{10} - i\Gamma_3)(2\omega - \omega_{20} - i\Gamma_3)},
\]

\[
\chi^{(3)}_{3\omega} = \frac{\varepsilon^2 \sigma_v M_{01} M_{12} M_{23} M_{30}}{\varepsilon_0 \hbar (\omega - \omega_{10} - i\Gamma_3)(2\omega - \omega_{20} - i\Gamma_3)(3\omega - \omega_{30} - i\Gamma_3)},
\]

The parameters used in Eqs. 6–8 are defined as: \( \sigma_v \) is a three-dimensional concentration of electrons involved in the transition, \( M_{ij} = \langle \psi_i(z) | e^z | \psi_j(z) \rangle \), \( i,j = 0,1,2,3 \) is the off-diagonal matrix element, \( \delta_{01} = | M_{00} - M_{11} |, \omega_{ij} = \frac{\hbar}{\hbar} \) is the transition frequency, and finally \( \Gamma_k = \frac{1}{T_k}, (k = 1,2,3) \) are damping terms associated with the lifetime of the electrons involved in the transitions.
3 Results and discussions

In this section, we present the outcome of our theoretical research on the GaAs QW with asymmetrical Gaussian potential. As previously commented, we consider both the influence of external static electric field ($F$) and magnetic field ($B$), whereas the application of a non-resonant ILF ($\alpha_0$) to the structure is also taken into account. The effects on the NOR, SHG, and THG coefficients of these external fields have been investigated. The simulation is performed in two steps. In the first one, the effective mass and the parabolic-band approaches are applied to obtain the energy eigenvalues and eigenfunctions of a single electron. In the second step, the coefficients of the nonlinear optical responses are evaluated. In general, the results of the simulation point at a critical role played by the applied external fields in controlling the amplitude and the position of the resonant peaks of the NOR, SHG, and THG coefficients. Both a blue-shift and a red-shift of the peak positions are observed with varying external fields.

At the outset, we need to specify certain input physical parameters: $L=200$ Å, $V_0=228$ meV, $m^*=0.067m_0$ (where $m_0$ is the free electron mass), $e=1.602 \times 10^{-19}$ C, $\hbar=1.056 \times 10^{-34}$ Js, $\sigma_v=3 \times 10^{22} m^{-3}$, $\epsilon_0=8.854 \times 10^{-12}$, $\Gamma_1=1.0THz$, $\Gamma_2=5.0THz$ and $\Gamma_3=7.0THz$. The choice of $V_0$ is made in such a way that the active well depth would reproduce the typical conduction band offset of an AlGaAs/GaAs heterostructure.

Figure 1 exhibits a schematic representation of the spatial variations of confinement potential profile together with wave functions corresponding to the first four lowest energy levels -depicted as the horizontal asymptotes for large enough values of the position along the growth direction. Dashed lines stand for the potential profile, subband energy levels, and related corresponding wave functions under $F=0$, $B=0$, and $\alpha_0=0$ conditions.

In Fig. 1a a static electric field is applied along the growth direction ($F=30$ kV/cm). The confinement potential of the structure undergoes a field-induced deformation, with the narrowing of the effective quantum well width ($L$). This causes a stronger spatial localization noticed as a shift of the probability density distributions towards the left-hand infinite potential barrier of the AGPQW. Thus, the subband levels position at higher energy. The ground state is less affected and slightly shifts upward. However, the higher electron states feel higher displacements. This external effect strengthens at higher electric field intensities. Consequently, the increment of $F$ causes the energy differences between the ground state and excited states to rise.

A similar scenario is depicted in Fig. 1b, where an in-plane oriented static magnetic field ($B=10$ T) is applied to the structure. Under the magnetic field, the system feels the
parabolic confinement potential, which is proportional to the square of the growth direction (constant $\times z^2$). Like the $F$ field, the $B$ field increases the degree of carrier confinement. The first four energy subbands shift to the higher energy values. The magnetic field changes the electron confinement with the rise of all energy levels and a blue shift of all transition energies.

In contrast to the electric and magnetic field influence on the AGPQW, the ILF has a different effect on the structure. In Fig. 1c, the reader may see the resulting features on the confining potential and lowest electron states for an intense laser, $\alpha_0 = 100$ Å ($F=0$ and $B=0$). ILF drastically deforms the conduction band profile. As a result, $L$ expands and the spatial localization diminishes. As a result, the electron energy levels descend towards lower values, and the difference of subband energy levels decreases as well, compared with their values at zero fields. The electron probability densities spread over a larger interval of positions on the right side of the potential.

Figure 2 presents the energy differences between the ground state ($E_0$) and the excited states (namely, $E_1$, $E_2$, $E_3$) as functions of (a) the external static electric field, (b) the static magnetic field, and (c) the laser dressing parameter ($\alpha_0$). As seen from Fig. 1a, the applied external electric field reinforces the spatial confinement, with the rise of the allowed energy levels which brings, as a consequence, the increment of the subband energy levels difference (see Fig. 2a). On the other hand, the applied magnetic field has an even stronger effect on the energy difference between the subband energy levels compared to the applied electric field due to the higher degree of induced confinement (see Fig. 2b). The subband energy differences show a faster pace of augmenting with the applied magnetic field. As noticed, the energy differences approach each other at the higher applied magnetic field. This indicates that for higher field intensities, the QW confinement tends to that of a typical parabolic profile.

The difference outcome, related to the applied ILF, is seen in Fig. 2c, where the ILF has a noticeable role in the evolution of intersubband energy difference values compared to those of the electric and magnetic fields. The figure exhibits that the energy differences initially decrease with the increment of the ILF, and this decrement continues up to 65 Å. Then the energy differences start increasing with a higher value of the oscillating ILF. The reason behind this oscillation can be attributed to the different rates of energy lowering shown by each of the levels considered. That is, above a certain value of the ILF strength, the fall in exciting energies becomes slower while the ground state one keeps its rate of descent more or less unchanged. Thus, the energy difference between the subband energy levels starts increasing again.

After investigating the effects of external fields on the potential profile and the lowest energy differences, we proceed to calculate the intersubband-related NOR, SHG, and THG.

![Fig. 2](image)

**Fig. 2** The energy difference between the ground state ($E_0$) and the first ($E_1$), second ($E_2$), and third ($E_3$) excited states as a function of the external **a** electric field **b** magnetic field **c** ILF
coefficients of the AGPQW in presence of applied external fields. Analyzing the optical rectification response, Fig. 3 presents NOR coefficient as a function of the incident photon energy under different external field conditions, considering a single field influence in each case. Figure 3a shows the sole effect of the static electric field ($F = 0$, 15, 30 kV/cm). We observe a blue shift of the peak positions, and the peak amplitude is decreased with the increment of $F$. The same approach is applied with the change of $B$ (0, 10, 20 T) in Fig. 3b. The external magnetic field also leads to a blue shift of the peak positions. The peak amplitude decreases with the increment of $B$. Finally, the laser dressing parameter ($\alpha_0$) is changed for 0, 50, 100 Å. The $\alpha_0$ leads to a red shift of the peak positions with a higher peak signal. The more $\alpha_0$ results decrease of the peak amplitude with a subsequent blue-shift compared to the lower value of $\alpha_0$. The NOR coefficient is mainly affected by the external value parameters. The peak positions are governed by the variation of $E_1-E_0$ as shown in Fig. 2. Meanwhile, the behavior of the peak amplitude can be understood by observing the changes in the involved dipole matrix element product appearing in the second column of Tables 1, 2, 3.

By using the same steps that we used in the simulation for Fig. 3, we obtain the SHG coefficient (see Fig. 4). The SHG spectrum is consistent with the features of the spectrum discussed in Fig. 3. $F$ and $B$ fields result in blue-shift with decreasing peak signal. However, the laser dressing parameters have an oscillation effect on the spectral distribution of SHG coefficients similar to the discussed in the case of varying electric field intensity. Again, the features of the resonant peak positioning are explained by the respective dependences of $E_1-E_0$ and $(E_2-E_0)/2$ depicted in Fig. 2, whilst the respective amplitudes behave according to the changes in the product of related dipole matrix elements shown in the third columns of Tables 1, 2, 3.

Finally, THG coefficients are spectrally resolved at different external fields ($F$, $B$, $\alpha_0$), as shown in Fig. 5. The peaks (under the effect of $F$ and $B$) are blue-shifted with decreasing peak amplitude. In addition, the oscillating ILF has a strong effect on the THG spectrum.
Note that the peak yield (for $\alpha_0 = 50 \text{ Å}$) in Fig. 5c is divided by a factor of 10 for visual comparison of the peaks. The ILF shifts the resonant peak to lower energy (red-shift), and then the higher intense laser field results in a resonant peak to shift higher energy (blue-shift). It is possible to observe that the amplitude of the THG coefficient is enhanced only in the case of the application of intense laser radiation (see the fourth column of Table 3). In the other two cases, the increment in the external probe intensity leads to a fall in magnitude, compared with the zero-field situation.
Reaching this point, it is worth commenting that ILF acts as an amplifier of the NOR, SHG, and THG nonlinear optical responses associated with main intersubband transitions in the AGPQW. In all cases, the application of this non-resonant signal produces an increment in the amplitudes of the respective coefficients. This has to do with the induced modification of the non-permanent dielectric polarization represented by the contributions of the off-diagonal dipole matrix elements between the states involved. Since these elements are directly related to the spatial extension of the states and the overlap between them, one may identify this growth in amplitude with the behavior exhibited by the allowed electron states as shown in Fig. 1c. Accordingly, a higher degree of spatial overlapping plus the greater extension of the electron wavefunctions -related to the effective widening of the QW- are responsible for the commented signal enhancement.

4 Conclusions

In this work, we theoretically studied the effect of non-resonant ILF, static electric, and magnetic fields on the NOR, SHG, and THG coefficients of AGPQW. The system is exposed to the external static electric field along the growth direction, in-plane oriented magnetic field, and the oscillating non-resonant ILF. The presented results could be used as the platform for real-time calculation and simulation and to solve physical problems. The simulation results have the potential to supplement a real-time experiment done in the laboratory. The calculations depend on the electron subband energy spectrum under the effective mass approximation and evaluation of the light-induced permanent and non-permanent dipole polarization. The simulation results for the AGPQW show that externally applied fields play a significant role in the shift of the transition energies together with the corresponding dipole moment matrix elements. The effect of these external fields induces either a reduction or an increment in the peak amplitude of the nonlinear coefficients, the latter being particularly associated with the presence of the intense non-resonant laser radiation.

The possible significance of the present study lies in the analysis of the behavior of a number of nonlinear optical properties under the influence of external fields. As is well known, the SHG and THG are great promise in optical microscopy and have been used for three-dimensional imaging of biological samples. For this reason, it might be of interest to experimental researchers working on the subject. Besides, the simulation technique provides an easy-to-implement method to study complex phenomena of nonlinear optical properties. It can be used to demonstrate nonlinear optics in a classroom setting, where the virtual experiments can be performed in real-time in front of the students.

Author contribution FU Numerical calculations, Writing of the article. AT Suggested the problem, Discussion of optical properties. MS Electronic structure discussion, Diagonalization method. MEMR Electronic structure discussion, Diagonalization method.

Data availability All data, figures, and the program are available. The corresponding author will provide all files upon request.

Declarations

Conflict of interest The authors declare that they have no conflict of interests.
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