Electric-field-induced Nonlinear Optical Rectification, Second- and Third-harmonic Generation in Asymmetrical Quantum Well

Serpil SAKIROGLU

1Dokuz Eylül University, Faculty of Science, Physics Department, 35390, İzmir

(Alınış / Received: 04.10.2016, Kabul / Accepted: 26.05.2017, Online Yayınlanma / Published Online: 17.06.2017)

Keywords
Morse potential, Electric field, Nonlinear optics, Quantum well

Abstract: The effects of a static electric field on the nonlinear optical rectification, second- and third-harmonic generation in a quantum well described by Morse potential are theoretically investigated. Analytical expressions for the optical coefficients due to intersubband optical transitions with an applied electric field are extracted from the compact-density matrix approach and iterative scheme. Numerical results presented for a typical GaAs quantum well reveal the feasibility of control of optical transitions between the size-quantized subbands. In addition, we have to emphasize the fact that the optical response of the system is remarkably sensitive to the electric field and structural range parameter.

1. Introduction

Over the last two decades, investigation of the characteristics of optical and electronic properties of semiconductor quantum wells (QWs), quantum wires, quantum dots and superlattices has become a fast-growing research area due to their potential applications in opto- and nanoelectronics [1–4]. In these structures, enhanced quantum confinement effects result in observation of unique nonlinear optical behavior very different from their bulk counterparts [5–7]. Moreover, the adjustability of the optical susceptibility by changing their size, shape, environmental conditions and external fields, makes these materials attractive both from theoretical and experimental point of view [8]. Among the nonlinear optical properties in nanostructures, nonlinear optical rectification coefficient (NOR), second-harmonic generation (SHG) and third-harmonic generation (THG) have been studied intensively [9–12]. Close relation between the asymmetry in confinement potential and second-order nonlinear optical properties in low-dimensional materials has been reported in diverse set of theoretical studies [13–18] which bring up the fact that even-order nonlinear optical responses are usually very small in symmetric quantum structures. Inversion symmetry in a potential profile can be broken simply by applying an electric field to a symmetric structure or by using both compositional grading and layer thickness variations [19, 20]. Advanced material growing techniques render possible the production of nanostructures with desired size, shape and chemical environment. Accordingly, considerable attention has been paid on the exploration of the optical properties of single and multiple QWs with semi-parabolic, semi-exponential, Pöschl-Teller, asymmetrical Gaussian, pseudoharmonic etc. potential profiles [3, 21–24].

Among different possible shapes for modeling of the confinement of quantum wells [25], owing to its asymmetrical structure Morse potential could be an appropriate to examine the nonlinear optical properties. Possessing an adjustable asymmetry stands as the most remarkable property of this potential. Gurnick and DeTemple first searched the nonlinearities in Morse quantum well and showed 10 to 100 times larger than the bulk materials [26]. Intersubband optical absorption and changes in
the index of refraction in this system have been examined by Feng-mei et al. [27, 28]. Besides, Refs. [29, 30] studied theoretically the influence of polaron on the change of refractive index and optical absorptions in a Morse quantum media. For the practical realization of the system under debate, we refer readers to Refs. [8, 26]. Very recently, Ref. [31] analyzed the combined effects of electric field and high-frequency intense laser field on the linear and third-order nonlinear optical absorption coefficients and refractive index changes.

However, to the best of our knowledge, effects of a static electric field on the optical harmonic generations in this system have not been investigated so far. Optical experiments on electric field-tunable quantum wells make manifest the electric field-controllability of the optical response of these structures [32]. An applied electric field modifies the potential energy profile of the structure and accordingly alters the subband energy levels [33]. Moreover, from the practical point of view, survey of the external field effect can provide an information for applications in optical switches. In this context, the main purpose of the present work is to expose in-depth research for a better understanding of the optical response of an asymmetrical quantum well. We address ourselves to survey the effects of well geometry and external static electric field on the NOR, SHG and THG in Morse quantum well. We have to remark that despite the system under debate is the same as in Ref. [31], the optical characteristics investigated in both papers are different.

The rest of the paper is organized as follows: Theoretical framework is briefly presented in Section 2. Section 3 is dedicated to the numerical results and finally, conclusions are given in Section 4.

2. Material and Method

Electrons in the Morse quantum well under the influence of an uniform static electric field, F, applied along the growth direction can be described within the framework of the effective mass approximation by the Hamiltonian:

\[ H = -\frac{\hbar^2}{2m^*} \frac{d^2}{dz^2} + V(z) + eFz, \]

where \(m^*\) is the electron effective mass, \(e\) is the absolute value the electron charge and \(z\) denotes the growth direction. The functional form of the Morse potential is given by [34, 35]

\[ V(z) = V_0 (1 - e^{-\alpha z})^2, \quad (-\infty < z < \infty) \]

where the parameter \(1/\alpha\) is related to the range and controls the asymmetry of the confining potential. Depth of the well is set to \(V_0 = \lambda^2E_0\) where \(E_0 = \hbar^2\alpha^2/2m^*\). In the absence of electric field, the finite number of bound eigenstates can be calculated via \(E_n = E_0 \left[ 2\lambda (n + 1/2) - (n + 1/2)^2 \right]\), where the threshold for the continuum is determined by an integer \(n = 0, 1, ... < \lambda - 1/2\) [36]. The most remarkable property of this potential is the parameter \(\lambda\) which controls the number of bound states. Since we focus on the analysis of the NOR, SHG and THG, in our work the parameter \(\lambda\) is taken to be 4. The four lowest electron states in electric field-tunable Morse quantum well have been calculated numerically via finite element method [37, 38].

In order to calculate the coefficients of NOR, SHG and THG, one considers that the system is irradiated by a light field with frequency \(\omega\) applied with polarization along the growth direction of the quantum well. Interaction of electromagnetic field with the system can be evaluated by means of the density-matrix approach and iterative procedure [39, 40]. As a consequence, the following expressions for the NOR, SHG and THG susceptibility are obtained [1, 9]

\[
\chi_0^{(2)} = \frac{4\hbar^3\rho_e^2 \mu_0^2 \delta_{01}}{\varepsilon_0 \hbar^2} \times \frac{\omega_{01}^2 (1 + \Gamma_2/\Gamma_1) + (\omega^2 + \Gamma_2^2)(\Gamma_2/\Gamma_1 - 1)}{(\omega_{01} - \omega)^2 + \Gamma_2^2} \left[ (\omega_{01} + \omega)^2 + \Gamma_2^2 \right],
\]

\[
\chi_{2ab}^{(2)} = \frac{e^3 \rho_e}{\varepsilon_0 \hbar^2} \mu_{01} \mu_{12} \mu_{20} \left( \omega - \omega_{01} - \Gamma_3 \right) \left( 2\omega - \omega_{20} - \Gamma_3 \right),
\]

\[
\chi_{3ab}^{(3)} = \frac{e^4 \rho_e}{\varepsilon_0 \hbar^3} \mu_{01} \mu_{12} \mu_{23} \mu_{30} \left( \omega - \omega_{01} - \Gamma_3 \right) \left( 2\omega - \omega_{20} - \Gamma_3 \right) \left( 3\omega - \omega_{30} - \Gamma_3 \right).
\]

Parameters used in equations are defined as: \(\delta_{01} = |\mu_{00} - \mu_{11}|\), \(\mu_{ij} = |\langle \psi_i | \psi_j \rangle| (i, j = 0, 1, 2, 3)\) is the off-diagonal matrix element when the eigenfunctions \(\psi_i(z)\) are obtained from numerical solution of Schrödinger equation corresponding to Eq.(1) by using the finite element method, and \(\omega_{ij} = (E_i - E_j)/\hbar\) is the transition frequency. In addition, \(\varepsilon_0\) is the vacuum permittivity and \(\rho_e\) is the electronic density. \(\Gamma_k = 1/\tau_k\) with \(k = (1, 2, 3)\) are damping terms associated with the lifetime of the electrons involved in the transitions.

3. Results

In this section, we will discuss the effects of electric field on the NOR, SHG and THG coefficients in a typical GaAs/AlGaAs Morse quantum well. The parameters adopted are: \(\rho_e = 5 \times 10^2\) m\(^{-3}\), \(m^* = 0.067m_0\), \(T_1 = 1.0\) ps, \(T_2 = 0.2\) ps and \(T_3 = 0.5\) ps. GaAs semiconductor materials are utilized in nonlinear optical applications because of an achievement of near-resonant excitation due to their fundamental bandgap in the optical region of electromagnetic spectrum [41]. Initially in Fig.1(a) we present the variation of the confinement potential for three different values of range parameter \(a\) considering the external electric field. Blue, green and red lines denote the range parameter \(a = 0.13869, 0.15726, 0.17385\) nm\(^{-1}\) corresponding to the well depths of \(V_0 = 175, 225, 275\) meV whereas solid, dashed and dotted lines represent the strengths of the electric fields of 0, 25, 50 kV/cm, respectively. The figure reveals that the asymmetry of the Morse quantum well decreases with the increase in the parameter \(a\). Besides, augmentation in the electric field causes a disruption in the asymmetry with narrowing the effective width of the potential. Electric field has prominent influence in larger asymmetry cases. Before
discussing the optical properties under interest, since it would beneficial to analyze the single-electron energies, in the Fig. 1(b) we present the variation of the first four lower-lying energy levels as a function of electric field. Highly accurate energy eigenvalues and eigenfunctions are obtained from the numerical solution of Eq. (1). As given in Fig. 1(b), at higher values eigenenergies are greater due to the enhancement in confinement potential. The pronounced feature of this figure is the monotonic increase in all the energies with the strengthening in the external electric field.

In Fig. 2(a)-(b), we plot the NOR as a function of the incident photon energy for three different values of the range parameter $a$ considering the zero- and non-zero-electric field cases, respectively. In this figure blue, green and red lines correspond to $a = 0.13869, 0.15726, 0.17385$ nm$^{-1}$, respectively. In zero-field case, with the deviation from the asymmetry in the potential profile, i.e. for higher range parameter values, amplitude of the NOR coefficient reduces considerably and its peak moves toward higher energies. Turning on the electric field, results in reduction of the resonant peak values whose magnitudes are related with geometrical factor $\mu_0^2 |\delta_1|$. It’s observed that the higher the range parameter is, the smaller the factor which gives rise to decrescent peak magnitudes. Moreover, as seen from Fig. 2(b), blue-shifting in the energies caused by the electric field can be attributed to the narrowing in the effective well width resulting with increment in the energy difference between two lowest-lying levels. In order to examine the effect of electric field-$a$ parameter relationship on the magnitude of NOR, the peak value of the NOR coefficient at resonance frequency $\omega = \omega_0$ is displayed in Fig. 2(c) as a function of electric field. We see that the peak amplitude of $\chi_0^{(2)}$ is a monotonous function of electric field and reduces considerably by $F$. Meanwhile, pronounced influence of electric field is clearer in the case of quantum well with higher asymmetry. Moreover, for sufficiently higher electric fields the peak maximums for all values of $a$ approach to each other.

Fig. 3(a)-(c) is dedicated to the SHG coefficient as a function of incident photon energy for Morse potential under electric field. Color labeling in the following figures is the same as in Fig. 2. In Fig. 3(a) and (b), second-harmonic generation coefficient in the absence and presence of electric field is shown for three values of the range parameter, respectively. As seen from the figure, for each curve of SHG two peaks emerge: the peak at lower (higher) energy corresponds to $\omega = \omega_0$ ($\omega = \omega_0/2$). Explanation of this feature can be based on the non-constant energy spacings between successive energy levels. Application of an electric field eventuates with the increment in the peak amplitude and shift in the position of the peaks. It is also clear that for larger range parameter $a$, $\chi_2^{(2)}$ coefficient takes smaller peak.
values. For better understanding of the field-induced changes in the amplitude of the maximum peak of SHG, the variation of the magnitude of the maximum resonance peak in SHG is plotted in Fig. 3(c). Non-monotonic behavior, increasing up to some threshold electric field and after which is followed by a steady variation, is more apparent in case of small structure parameter. We should note that, the second-harmonic generation coefficient is affected strongly by the structure parameter and electric field manifests its effect by inducing a blue-shift in the resonant peaks.

The THG susceptibility as a function of the incident photon energy $h\omega$ is plotted for three values of the range parameter taking into account the electric field of 0 kV/cm and 25 kV/cm as seen in Fig. 4(a) and (b), respectively. The resonant peaks in THG are a consequence of the presence of three factors with form $[n \hbar \omega - E_{30} - i \hbar \Gamma_3]$ appearing in Eq. (5) and are a signature of unequal energy spacings. The first resonance emerges when the incident optical energy is equal to $E_{10}$ whereas second (third) peak is result of resonance at $E_{20}/2$ ($E_{30}/3$). Strengthening in electric field value produces a blue-shift in the spectrum of which explanation is merely related to the electric field acting as a confining agent. Nevertheless, the absolute value of THG increases noticeably when the structure parameter goes from $a = 0.17385$ to $a = 0.13869$. Besides, as seen from Fig. 4(c) the maximum peak value of the THG as a function of external electric field shows similar behavior as SHG. Electric-field induced changes in the magnitude of the THG peaks are related with the corresponding dipole matrix elements appearing in Eq. (5). Enhancement of the factor $\mu_{01}\mu_{12}\mu_{23}\mu_{30}$ with increasing field is observed for all $a$ values, but especially for the smallest range parameter it has more pronounced influence. We note that, as higher the asymmetry, as larger the peak values of THG. Another feature that is obvious from this figure is that rather significant variation with the increase of the field is observed for the smallest range parameter.

In the light of the aforementioned observations it can be said that with a proper optimization of range parameter and electric field, designated optical characteristics in quantum well can be achieved.

4. Discussion and Conclusion

In this paper, nonlinear optical rectification, second- and third-harmonic generation in Morse quantum wells are studied with an emphasis mainly focused on the influence of external static electric field. The results show that the magnitude and the positions of the resonance energies of

Figure 3. (color online) The $\chi^{(2)}_{220}$ as a function of the photon energy (a) the absence and (b) presence of electric field for varying values of range parameter. (c) Variation of the peak magnitude of the maximum resonance peak of SHG coefficient with respect to electric field.

Figure 4. (color online) The $\chi^{(3)}_{320}$ as a function of the photon energy in the (a) absence and (b) presence of electric field for different values of range parameter. (c) Variation of the maximum of the pronounced resonance peak of THG coefficient with respect to electric field.
the coefficients under debate are strongly sensitive to the strength of electric field and the structural parameter governing the confinement potential. Increasing electric field values shift the maximum peaks of NOR, SHG and THG toward higher-energy regions while cause a reduction in NOR amplitudes. Enhancement in the asymmetry of the quantum well evoke an increase in SHG and THG susceptibility. Large nonlinearities associated with intersubband optical transitions can be obtained by a proper choice of structural parameter of Morse quantum well. It is expected that the results of our paper can be useful not only in the elucidation of the physical properties of the system but also in the practical realization of quantum wells with interesting optoelectronic properties.

Acknowledgment

The author would like to thank to Prof. Dr. Ismail Sökm-en for his valuable suggestions and critical reading of the manuscript.

References


