Electric field effect on the second-order nonlinear optical properties in semiparabolic quantum wells

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Abstract

Electric field effect on the second-order nonlinear optical properties in semiparabolic quantum wells are studied theoretically. Both the second-harmonic generation susceptibility and nonlinear optical rectification depend dramatically on the direction and the strength of the electric field. Numerical results show that both the second-harmonic generation susceptibility and nonlinear optical rectification are always weakened as the electric field increases where the direction of the electric field is along the growth direction of the quantum wells, which is in contrast to the conventional case. However, the second-harmonic generation susceptibility is weakened, but the nonlinear optical rectification is strengthened as the electric field increases where the direction of the electric field is against the growth direction of the quantum wells. Also it is the blue (or red) shift of the resonance that is induced by increasing of the electric field when the direction of the electric field is along (or against) the growth direction of the quantum wells. Finally, the resonant peak and its corresponding to the resonant energy are also taken into account.

Keywords: Quantum wells; Electronic states; Nonlinear optics

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1. Introduction

In the past few years, the study of the optical properties in semiconductor quantum wells (**QWs**) have been so intensively studied [1, 2, 3, 4, 5]. This is because the nonlinear effects can be enhanced more dramatically in **QWs** than in bulk materials, which can provide a promising route to the fabrication of semiconductor quantum micro-device, such as high-speed electro-optical modulators, far-infrared photo detectors, and semiconductor optical amplifiers and so on [6, 7, 8, 9]. For the bulk susceptibility, the nonlinear effect is not very large because of the symmetry of the crystal structure. For nano materials also with symmetric structure, even-order nonlinear optical effects are usually vanish in theory. Thus the contributions to the second order nonlinear optical susceptibilities are zero for a symmetrical **QW**, but as the symmetry is broken, nonvanishing contributions to second order nonlinear optical susceptibilities are expected to appear[2]. Consequently, in order to obtain the enhanced second order nonlinear optical susceptibilities in **QWs**, externally applied electric fields are used to remove the symmetry [1, 2, 3] or the **QWs** structures are produced with a built-in asymmetry using advanced material growing technology [4, 5].

Among the nonlinear optical properties, it is attracted much attention to the second order nonlinear optical properties, such as optical rectification (**OR**) and second-harmonic generation (**SHG**). It is because the second-order nonlinear processes are the simplest and the lowest-order nonlinear effects, and the magnitudes of these second-order nonlinear coefficients are usually stronger than that of the higher-order ones, as the symmetry of quantum systems is broken. For example, Karabulut and Baskoutas studied the second and third harmonic generation susceptibilities for the case of spherical quantum dot with parabolic confinement subjected to an external electric field with the presence of an impurity. Their results indicate an increase of the electric field can enhance the peak values of the second[10]. In 2003, Zhang and Xie [1] reported nonzero contributions to **SHG** susceptibilities for both parabolic and semiparabolic **QWs**. However, those pertaining to parabolic **QWs** are completely wrong and contradicts with well-established literature, which had been commented by Karbulut et al[2]. Karabulut and Safak later[11] studied the nonlinear **OR** in semiparabolic **QWs** with an applied electric field. The nonlinear optical properties in the semiparabolic **QW** have been attracted much attention, such as exciton effect[12], optical absorption[13] and **SHG**[14]. Recently, Guo and Du [15] have reported their

results for linear and nonlinear optical absorption coefficients and refractive index changes in asymmetrical Gaussian potential **QWs** with applied electric field in the limit $z \ll L$ replaced the potential $-V_0 \exp(-z^2/2L^2)$ with the semiparabolic potential $-V_0(1 - z^2/2L^2)$. We find both the energy and the corresponding wavefunction for the low-lying state are wrong to use in works above [16, 17]. Unfortunately, these have not been attracted considerable attention by these authors[18, 19, 20]. After the other optical properties in the asymmetrical Gaussian potential **QWs** are investigated, such as nonlinear **OR** [18], **SHG** [19] and nonlinear optical absorption via two-photon process[20]. Factually, the direction of the electric field is of importance for studying the nonlinear effect. But there is little literatures for reporting it. Therefore, it is very necessary to investigate the nonlinear optical properties for electron confined in the semiparabolic **QWs** in the presence of the applied electric field where the direction of the electric field is along (or against) the growth direction of the **QWs**.

In this paper, electric-field-induced **SHG** susceptibility and nonlinear **OR** coefficient in semiparabolic **QWs** are investigated theoretically. We find that both the **SHG** susceptibility and nonlinear **OR** depend dramatically on the direction and the strength of the electric field. Numerical results show that the **SHG** susceptibility is always weakened as the electric field increases no matter the direction of the electric field is along (or against) the growth direction of the **QWs**. However, the nonlinear **OR** is weakened (or strengthen) as the electric field increases where the direction of the electric field is along (or against) the growth direction of the **QWs**. Also it is the blue (or red) shift of the resonance that is induced by increasing of the electric field when the direction of the electric field is along (or against) the growth direction of the **QWs**. Finally, the resonant peak and its corresponding to the resonant energy are also taken into account. This paper is organized as follows: Hamiltonian, the relevant wave functions and energy levels are briefly described in Section 2. Also the analytical expressions of the **SHG** susceptibility and **OR** coefficient in semiparabolic **QWs** are presented in this section. Numerical calculations and detailed discussions for typical $\text{Al}_x\text{Ga}_{1-x}\text{Al}/\text{GaAs}$ materials are given in Section 3. Finally, a brief summary is presented in Section 4.

2. Theory

Within the framework of effective-mass approximation, the Hamiltonian of an electron confined in semiparabolic **QWs** in the presence of electric field

along the z axis can be written by

$$H = -\frac{\hbar^2}{2m_e^*} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) + V(z) + q\eta Fz. \quad (1)$$

with

$$V(z) = \begin{cases} \frac{1}{2}m_e^*\omega_0^2 z^2, & z \geq 0, \\ \infty, & z < 0, \end{cases} \quad (2)$$

where, z represents the growth direction of the **QWs**. m_e^* is the effective mass in materials. $\hbar$ is the Planck constant. ω_0 is frequency of the semi-parabolic confined potential in **QWs**, F is the strength of the electric field, $\eta = +1$ (or -1) describes the direction of the electric field is along (or against) the growth direction of the **QWs** and q is the absolute value of the electric charge. Under the envelope wave-function approximation, the eigenfunctions $\Psi_{t_n,k}(r)$ and eigenenergies $\varepsilon_{t_n,k}$ are the solutions of the Schrödinger equation for H and are given by [1, 19, 21, 22]

$$\Psi_{t_n,k}(r) = \Phi_{t_n}(z) U_c(\mathbf{r}_{\parallel}) \exp(i\mathbf{k}_{\parallel} \cdot \mathbf{r}_{\parallel}) \quad (3)$$

and

$$\varepsilon_{t_n,k} = E_{t_n} + \frac{\hbar^2 k_{\parallel}^2}{2m_e^*} \quad (4)$$

Here, $\mathbf{k}_{\parallel}$ and $\mathbf{r}_{\parallel}$ are the wave vector and coordinate in the xy plane and $U_c(r)$ is the periodic part of the Bloch function in the conduction band at $\mathbf{k} \equiv 0$. $\Phi_{t_n}(z)$ and E_{t_n} can be obtained by solving the following Schrödinger equation

$$\begin{aligned} H_z \Phi_{t_n}(z) &= \left[-\frac{\hbar^2}{2m_e^*} \frac{\partial^2}{\partial z^2} + V(z) + q\eta Fz \right] \Phi_{t_n}(z) \\ &= E_{t_n} \Phi_{t_n}(z). \end{aligned} \quad (5)$$

The electronic energy levels and corresponding wave functions are given as follows [1]

$$E_{t_n} = (2t_n + 1 - \alpha^2 \beta^2) \frac{\hbar \omega_0}{2}, n = 1, 2, 3, \dots, \quad (6)$$

and

$$\Phi_{t_n}(z) = N_n \exp(-\alpha^2(z + \beta)^2) H_{t_n}(\alpha(z + \beta)), \quad (7)$$

with

$$\alpha = \sqrt{\frac{m_e^* \omega_0}{\hbar}}, \beta = \frac{q\eta F}{m_e^* \omega_0^2} \quad (8)$$

where H_{t_n} is the Hermite functions and t_n is real, N_n is the normalization constant. t_n is determined by $\Phi_{t_n}(z = 0) \equiv 0$, that is to say, the relation always should be satisfied as $H_{t_n}(\alpha\beta) \equiv 0$. Obviously, $t_n = 2n + 1$ as the electric field is in absence, where $n = 0, 1, 2 \dots$.

The formulas of the **SHG** susceptibility and the **OR** coefficient in the two models will be derived by using the compact-density-matrix method and the iterative procedure. The system is excited by electromagnetic field $\mathbf{E}(t) = \tilde{E}e^{i\omega t} + \tilde{E}e^{-i\omega t}$. Let us denote ρ as the one-electron density matrix for this regime. Then the evolution of density matrix is given by the time-dependent Schrödinger equation

$$\frac{\partial \rho_{ij}}{\partial t} = \frac{1}{\hbar} [H_0 - qz\mathbf{E}(t), \rho]_{ij} - \Gamma_{ij}(\rho - \rho^{(0)})_{ij}. \quad (9)$$

where H_0 is the Hamiltonian for this system without the electromagnetic field $\mathbf{E}(t)$; $\rho^{(0)}$ is the unperturbed density matrix; Γ_{ij} is the relaxation rate.

Eq.(9) is calculated by the following iterative method [1]

$$\rho(t) = \sum_n \rho^{(n)}(t), \quad (10)$$

with

$$\begin{aligned} \frac{\partial \rho_{ij}^{(n+1)}}{\partial t} &= \frac{1}{i\hbar} \{ [H_0, \rho^{(n+1)}]_{ij} - i\hbar \Gamma_{ij} \rho_{ij}^{(n+1)} \} \\ &- \frac{1}{i\hbar} [qz, \rho^{(n)}]_{ij} \mathbf{E}(t). \end{aligned} \quad (11)$$

The electric polarization of the quantum system due to $\mathbf{E}(t)$ can be expressed as

$$P(t) \approx \varepsilon_0 \chi_\omega^{(1)} \tilde{E} e^{i\omega t} + \varepsilon_0 \chi_{2\omega}^{(2)} \tilde{E}^2 e^{2i\omega t} + c.c. + \varepsilon_0 \chi_0^{(2)} \tilde{E}^2, \quad (12)$$

where $\chi_\omega^{(1)}$, $\chi_{2\omega}^{(2)}$ and $\chi_0^{(2)}$ are the linear, **SHG** and optical rectification susceptibility, respectively.

With the same compact density matrix approach and the iterative procedure as [23, 24], the analytical expressions of the **SHG** susceptibility and the **OR** coefficient are given as [1, 2, 11, 19, 25]

$$\begin{aligned}\chi_{2\omega}^{(2)} &= \frac{q^3 \sigma M_{12} M_{23} M_{31}}{\varepsilon_0} \\ &\times \frac{1}{(E_{31} - 2\hbar\omega + i\hbar\Gamma_0)(E_{21} - \hbar\omega + i\hbar\Gamma_0)}\end{aligned}\quad (13)$$

and

$$\begin{aligned}\chi_0^{(2)} &= \frac{q^3 \sigma M_{12}^2 \delta_{12}}{\varepsilon_0 \hbar^2} \\ &\times \frac{2E_{21}^2}{[(E_{21} - \hbar\omega)^2 + \Gamma_0^2][(E_{21} + \hbar\omega)^2 + \Gamma_0^2]}\end{aligned}\quad (14)$$

where σ is the surface density of electrons in the **QWs**, Γ_0 is the phenomenological relaxation rate, E_{ij} is the energy interval of two different electronic states, $\delta_{ij} = \langle j|z|j\rangle - \langle i|z|i\rangle$ and M_{ij} is the off-diagonal matrix element which is given by $M_{ij} = \langle i|z|j\rangle$ where $(i, j = 1, 2, 3)$.

3. Results and Discussions

In this section, the electric-field-induced **SHG** susceptibility and **OR** coefficient in semiparabolic quantum wells are investigated theoretically. The parameters adopted in the present work are as follows: $m_e^* = 0.067m_0$ (m_0 is the electron mass), the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz, $\sigma = 5.0 \times 10^{24} \text{ m}^{-3}$, and $\Gamma_0 = 1/0.14 \text{ ps}$ [1, 2, 11].

The peak of the **SHG** susceptibility $\chi_{2\omega}^{(2)}$ is plotted as a function of the electric field strength F and the resonant energy E_{res} with the confined potential frequency $\omega_0 = 3.6 \times 10^{14} \text{ Hz}$ for $\eta = +1$ in Fig.1 and $\eta = -1$ in Fig.2. We note that it is wrong to calculate the peaks for the **SHG** susceptibility $\chi_{2\omega}^{(2)}$ in Ref.[1]. This will not generally be the case because the condition of two-photon resonance is only satisfied in the absence of the electric field. Zhang and Xie [1] concluded that there is one peak of $\chi_{2\omega}^{(2)}$ and the value of $\chi_{2\omega, max}^{(2)}$ increase with the increase of the applied electric field F , where F

varies from 0 to 10.0×10^7 V/m. The results seem to be right and conform to the theory of the applied electric field enhanced the **SHG** susceptibilities, but in fact it is wrong. Our results are obvious different from that reported in Ref.[1]. For both cases, we find that (1) a transition from an approximate two-photons resonance to two single-photon resonances will appear when the electric field strength is more than a certain value: 8.0×10^7 V/m for $\eta = 1$ [see in Fig.1] and 9.0×10^7 V/m for $\eta = -1$ [see in Fig.2]; (2) The main (or weak) peak decreases as the electric field increases in the two cases and the effect is more obvious for $\eta = 1$ than that for $\eta = -1$; (3) The resonant energy E_{res} related to the weak peak is larger (or smaller) than that related to the main peak for $\eta = 1$ (or $\eta = -1$); (4) The blue (or red) shift of the resonance can be induced by increasing of the electric field for $\eta = 1$ (or $\eta = -1$) (5) The **SHG** susceptibility obtained in this system can reach the magnitude of 10^{-6} m/V;

To better understand the effect of the electric field on the **SHG** susceptibility in semiparabolic **QWs**, the **SHG** susceptibility $\chi_{2\omega}^{(2)}$ is plotted in Fig.3 ($\eta = 1$) and Fig.4 ($\eta = -1$) as a function of the photon energy E_p with four different strengths of the electric field $F = 0, 2.0 \times 10^7, 5.0 \times 10^7$ and 10.0×10^7 V/m for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz. We find that the **SHG** susceptibility will decrease with the increase of the electric field as reported by Ref.[21] (see in the Fig.6 of Ref.[21]), which is contrast to the conventional case as reported by Refs.[1, 3]. It is easily seen that the product of $M_{21}M_{32}M_{31}$ always decreases for $\eta = 1$ but always increases for $\eta = -1$ as the electric field increases. Obviously, the product of $M_{21}M_{32}M_{31}$ can not be used to interpret the reason why **SHG** susceptibility will decrease with the increase of the electric field as reported in Refs[1, 19]. The reason is that the condition of the two-photon resonance is not satisfied as the electric field is applied, i.e. $E_{21} \neq E_{31}/2$. And the difference between E_{21} and $E_{31}/2$ is more and more large as the electric field increases. That is to say, with the increase of electric field, resonant energy E_{res} obviously deviate from two-photon resonance condition; (2) It is the blue (or red) shift of resonance of photon that is induced as the strength of electric field increases because the energy spacing of the E_{21} and $E_{31}/2$ are increasing (or decreasing) with the increase of electric field for $\eta = 1$ (or $\eta = -1$); (3) A transition from an approximate two-photons resonance to two single-photon resonances will appear adjusted by the electric field. When the energy spacing of E_{21} and $E_{31}/2$ differ too much, the approximate condition of the two-photon resonance is not satisfied because the condition of approximation, i.e. $E_p \approx E_{21} \approx E_{31}/2$,

is not satisfied any more. However, the single photon resonance condition is easy to meet, so a transition from a two-photons resonance to two single-photon resonances will appear as the electric field increases; (4) For both cases, the single peak of the **SHG** susceptibility corresponding to the two-photons resonance occurs for the weak electric field about the photon energy $\hbar\omega \approx E_{21} \approx E_{31}/2$. Also the main and the weak peaks for the two single-photon resonances are always related to the resonant energy $E_{res} \approx E_{31}/2$ and $E_{res} \approx E_{21}$, respectively; (5) The line width of photon spectrum in the two cases ($\eta = \pm 1$) become more wider with the increase of the electric field because of the large of energy spacing of E_{21} and $E_{31}/2$ for a large electric field.

In Fig.5 ($\eta = 1$) and Fig.6 ($\eta = -1$), the peak of the **OR** coefficient $\chi_0^{(2)}$ is plotted as a function of the electric field strength F and the resonant energy E_{res} for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz. It is observed that a nonvanishing contributions to **OR** coefficient can arrive to the magnitude of 10^{-6} m/V even in the absence of the electric field because of the self-asymmetry of the semiparabolic QWs. To surprise to us, the peak of the **OR** coefficient $\chi_0^{(2)}$ decreases (or increases) almost linearly with the increasing of the electric field for $\eta = 1$ (or $\eta = -1$). Also the peak of the **OR** coefficient suffers a blue (or red) shift as the electric field increases for $\eta = 1$ (or $\eta = -1$). It is easily seen that the magnitude of the peak of the **OR** coefficient is always larger than that of the **SHG** susceptibility. Thus, the **OR** coefficient is more easily observed in the experiment for the **QWs** especially in the presence of the electric field. Furthermore, only single peak of the **OR** coefficient $\chi_0^{(2)}$ is observed because of the single photon resonance condition, which is different from the peak of the **SHG** susceptibility.

The **OR** coefficient $\chi_0^{(2)}$ versus the photon energy E_p is shown in Fig.7(a) with four different strengths of the electric field $F = 0, 2.0 \times 10^7, 5.0 \times 10^7$ and 10.0×10^7 V/m for $\eta = 1$. As the same to the Fig.5, it is easily observed in Fig.7(a) that the peak of the **OR** coefficient $\chi_0^{(2)}$ decreases and a blue shift of resonant peak is induced as the electric field increases. In order to interpret these prosperities, the geometric factor δ_{ij} are plotted in Fig.7(b) as a function of the electric field strength for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz. Obviously, the peak of the **OR** coefficient $\chi_0^{(2)}$ is proportional to the geometric factor δ_{ij} . It is easily seen that the geometric factor δ_{ij} decreases as the electric field increases. As we all know, when the condition of the single photon for the **OR** coefficient (see in Eq.(14)), i.e.

$E_p \equiv E_{21}$, is satisfied, a peak of the **OR** coefficient will appear. It is easily seen that the energy spacing E_{21} is decreasing with increasing of the electric field (see the inset of the fig.3(b)). Thus it is the blue shift of the resonance that is induced with the increase of the electric field strengths. In Fig.8, the **OR** coefficient $\chi_0^{(2)}$ versus the photon energy E_p is shown with four different strengths of the electric field $F = 0, 2.0 \times 10^7, 5.0 \times 10^7$ and 10.0×10^7 V/m for $\eta = -1$. The results of the Fig.8 are obviously different from that of the Fig.7. It is easily seen that the peak of the **OR** coefficient $\chi_0^{(2)}$ increases and a red shift of resonant peak is induced as the electric field increases. Also the peaks of $\chi_0^{(2)}$ for $\eta = -1$ are obvious larger than that for $\eta = 1$.

4. Conclusion

In summary, the effect of the electric field on the **SHG** susceptibility and the nonlinear **OR** coefficient in semiparabolic **QWs** have been studied in detail. As we know, with the recent advances in nano fabrication technology, it is possible to produce the manufactures of such the semiparabolic **QWs**. Numerical results show that both the **SHG** susceptibility and nonlinear **OR** are always weakened as the electric field increases where the direction of the electric field is along the growth direction of the **QWs**, which is in contrast to the conventional case. However, the **SHG** susceptibility is weakened, but the nonlinear **OR** is strengthen as the electric field increases where the direction of the electric field is against the growth direction of the **QWs**. Also it is the blue (or red) shift of the resonance that is induced by increasing of the electric field when the direction of the electric field is along (or against) the growth direction of the **QWs**. Our theoretical results may make a contribution to experimental studies and provide a kind of approximative modeling for the practical application such as optoelectronics devices and optical communication, etc.

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Figure Captions

Figure 1 The peak of the **SHG** susceptibility $\chi_{2\omega}^{(2)}$ as a function of the electric field strength F and the resonant energy E_{res} for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz and $\eta = +1$.

Figure 2 The peak of the **SHG** susceptibility $\chi_{2\omega}^{(2)}$ as a function of the electric field strength F and the resonant energy E_{res} for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz and $\eta = -1$.

Figure 3 The **SHG** susceptibility $\chi_{2\omega}^{(2)}$ versus the photon energy E_p for $\eta = +1$ with four different strengths of the electric field $F = 0, 2.0 \times 10^7, 5.0 \times 10^7$ and 10.0×10^7 V/m. The product of $M_{21}M_{32}M_{31}$ of matrix elements M_{ij} in the inset (a) and the energy space ΔE of E_{21} and $E_{31}/2$ in the inset (b) are plotted as a function of the electric field strength for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz, respectively..

Figure 4 The **SHG** susceptibility $\chi_{2\omega}^{(2)}$ versus the photon energy E_p for $\eta = -1$ with four different strengths of the electric field $F = 0, 2.0 \times 10^7, 5.0 \times 10^7$ and 10.0×10^7 V/m (a). The product of $M_{21}M_{32}M_{31}$ of matrix elements M_{ij} (b) and the energy space ΔE of E_{21} and $E_{31}/2$ (c) are plotted as a function of the electric field strength for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz, respectively.

Figure 5 The peak of the **OR** coefficient $\chi_0^{(2)}$ as a function of the electric field strength F and the resonant energy E_{res} for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz and $\eta = +1$.

Figure 6 The peak of the **OR** coefficient $\chi_0^{(2)}$ as a function of the electric field strength F and the resonant energy E_{res} for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz and $\eta = -1$.

Figure 7 The **OR** coefficient $\chi_0^{(2)}$ versus the photon energy E_p for $\eta = +1$ with four different strengths of the electric field $F = 0, 2.0 \times 10^7, 5.0 \times 10^7$ and 10.0×10^7 V/m (a). Geometric factor δ_{ij} are plotted as a function of the electric field strength for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz (b).

Figure 8 The **OR** coefficient $\chi_0^{(2)}$ versus the photon energy E_p for $\eta = -1$ with four different strengths of the electric field $F = 0, 2.0 \times 10^7, 5.0 \times 10^7$ and 10.0×10^7 V/m (a). Geometric factor δ_{ij} are plotted as a function of the electric field strength for the confined potential frequency $\omega_0 = 3.6 \times 10^{14}$ Hz (b).

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