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Polaron effects on the optical rectification in electric-field-biased parabolic quantum wells

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Abstract. Second-order susceptibilities are calculated for weak-coupling optical polaron systems in the electric-field-biased parabolic quantum well structure. Most emphasis is devoted to the electron–LO-phonon interaction. The origin of the large optical rectification lies in the possibility of tuning independently the parabolic potential shape (force constant D in the potential is tunable) and the applied bias field F. The theoretical value of the optical rectification in this structure is more than a factor of 10 higher than in the structure where the electron–LO-phonon interaction is ignored. It has been shown that our results are independent of the electron–electron interaction and of the number of electrons in the parabolic quantum well.

1. Introduction

In the last two decades much attention has been focused on the study of polarons of reduced dimensionality in the context of the quantum-well-confined semiconductor structures. Of particular interest is the quasi-two-dimensional (Q2D) optical polaron, with most emphasis devoted to its strict two-dimensional (2D) characterization within the framework of an idealized approximation, accounting for the almost planar aspect of an electron in a thin quantum well and yet interacting with the bulk LO phonon modes of the well material [1, 2]. The common theoretical prediction of the relevant work in the literature [1–5] is that the electron interacts more effectively with the phonons in two dimensions and consequently certain polaron quantities scale by rather large factors over their corresponding bulk values.

Wide parabolic quantum wells have recently been grown [11] by tailoring the conduction-band edge of a graded $Al_xGa_{1-x}As$ semiconductor. In comparison to square QWs, parabolic quantum wells (PQWs) have been shown to exhibit properties such as a nearly uniformly spaced density of states for the electrons and holes [6]. It is expected that the optical nonlinearities are more sensitive to non-square quantum well shapes than to square well shapes [7]. Quantum confinement of carriers in a semiconductor parabolic quantum wells leads to the formation of discrete energy levels and the drastic change of optical susceptibilities [8]. One of the most remarkable properties of these quasi-two-dimensional electronic systems is that optical transition between the size-quantized subbands are feasible. The electrons are quantized into subbands where their wave functions in the growth direction have the form of envelope functions with an extension equal to the effective well width, i.e. in the few-nanometre range. Electromagnetic waves may induce electronic transitions between these subbands. The dipole matrix elements associated with

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these intersubband transitions have the same order of magnitude as the effective well width leading to extremely large absorption. The dipole matrix elements are thus in the nanometre range instead of the few picometres obtained in usual molecular or ionic system [9]. As even-order susceptibilities vanish in structures with inversion symmetry, finite second-order susceptibilities can only be observed if the symmetry of the conduction-band potential is broken through either the growth of an asymmetric well or the application of an external bias field. The optical non-linearities in parabolic quantum wells with an applied electric field have been studied [10]. However, the polaron effects have never been considered in these papers. We report here on the calculations of optical nonlinearities of weak-coupling optical polarons in electric-field-biased parabolic quantum well media, with most emphasis devoted to the influence of electron–LO-phonon interaction on optical susceptibilities. As we know, there has been little work on the inclusion of polaron effects when calculating the nonlinear susceptibility or even the linear susceptibility in low-dimensional structures, although several authors [18–22] have calculated the polaron energy and impedance function and have shown that they scale by as much as a factor of 10 in two dimensions compared with bulk materials. It is interesting and important that our calculations show that this is true for the optical rectification coefficient. That means the polaron effect plays an important role in the calculation of the optical rectification coefficient for a parabolic quantum well in an electric field.

It should be noted that for the present we take the confined electron as interacting with the bulk phonon modes only. Since in the most commonly studied compound materials (such as GaAs, for instance) the electron–phonon coupling is rather weak, an appropriate approach is to treat the Fröhlich interaction as a perturbation. In the following we restrict our considerations solely to the weak-coupling regime.

2. Theory

For simplicity, we will use units for which $2m^* = \hbar = \omega_{LO} = e = 1$ (m^* is the effective electron band mass, $\hbar$ is Planck's constant, ω_{LO} is the LO phonon frequency, and e is the electronic charge). The interactions between electrons in the conduction band can be separated into a Hartree term due to the electrostatic potential of the total electron density and an exchange–correlation term [15]. The exchange–correlation part of the ground-state energy can be described as a functional of the electron density [16]. It should be noted that our theory is limited to very low electron densities. In the local density approximation (LDA) this functional is assumed to have only local dependence on the electron density. Making this local approximation leads to a one-body Schrödinger-type equation, where the electrons move in a potential that is the sum of the external (parabolic) potential, the Hartree potential, and an exchange–correlation potential. The Hamiltonian describing the weak-coupling optical polaron systems in electric-field-biased parabolic quantum well media is given by

$$H = H_e + H_{ph} + H_{e-ph} \tag{1}$$

where

$$H_e = P^2 + P_z^2 + V(z) + V_H(z) + V_{xc}(z) + Fz \tag{2}$$

is the electron part,

$$H_{ph} = \sum_Q a_Q^+ a_Q \tag{3}$$

is the phonon part, and

$$H_{e-ph} = \sum_{Q} V_Q[a_Q \exp(i\boldsymbol{q}\cdot\boldsymbol{\rho})\exp(iq_z z) + \text{HC}]. \tag{4}$$

is the Fröhlich interaction. In the above, $(\boldsymbol{\rho}, z)$ and $(\boldsymbol{P}, P_z)$ denote the electron position and momentum, F is the external bias electric field. The interaction amplitude is related to the electron–phonon coupling constant α and the phonon wavevector $\boldsymbol{Q} = (\boldsymbol{q}, q_z)$ through $V_Q = (4\pi\alpha)^{1/2}/Q$.

The unperturbed wave equation for the electron is given by

$$H_e\Phi_{k,n}(\boldsymbol{\rho}, z) = \varepsilon_n(k)\Phi_{k,n}(\boldsymbol{\rho}, z) \qquad n = 0, 1, 2, \ldots \tag{5}$$

where

$$\Phi_{k,n}(\boldsymbol{\rho}, z) = \varphi_n(z)\phi_k(\boldsymbol{\rho}). \tag{6}$$

We have

$$(P_z^2 + V(z) + V_H(z) + V_{xc}(z) + Fz)\varphi_n(z) = e_n\varphi_n(z). \tag{7}$$

In this expression, $V(z) = \frac{1}{4}\omega_0^2 z^2 = \frac{1}{2}Dz^2$ is the parabolic potential, and $\omega_0 = (D/m^*\omega_{\text{LO}}^2)^{1/2}$ (D is the force constant in the potential, which will be treated as a tunable parameter referring to the degree of confinement in the z direction) stands for the dimensionless measure of the degree of confinement in the longitudinal (z) direction; $V_{xc}(z)$ is the exchange–correlation potential [17]:

$$V_{xc}(z) = -0.985\frac{1}{\varepsilon_0}n^{1/3}(z)\left(1 + \frac{0.034}{a_B^* n^{1/3}(z)}\ln[1 + 18.376a_B^* n^{1/3}(z)]\right) \tag{8}$$

where $a_B^* = 2\varepsilon_0$ (ε_0 is the vacuum permittivity); $V_H(z)$ is the Hartree term due to the electrostatic interaction of the electrons with themselves,

$$\frac{\mathrm{d}^2V_H(z)}{\mathrm{d}z^2} = -\frac{4\pi}{\varepsilon_0}[n(z) - N_D(z)] \tag{9}$$

where $N_D(z)$ is the density of positive charge necessary to maintain charge neutrality. The electron density $n(z)$ is given by

$$n(z) = \sum_i n_i(z) \tag{10}$$

$$n_i(z) = \frac{1}{2\pi}(\varepsilon_F - \varepsilon_i)|\varphi_i(z)|^2\Theta(\varepsilon_F - \varepsilon_i) \tag{11}$$

where $n_i(z)$ is the contribution of the ith subband to the charge density, $\Theta(\varepsilon)$is the Heaviside unit-step function ($\Theta = 0$ for $\varepsilon < 0$ and $\Theta = 1$ for $\varepsilon > 0$), and ε_F is the Fermi energy obtained from the condition

$$n_s = \sum_i N_i \tag{12}$$

$$N_i = \frac{1}{2\pi}(\varepsilon_F - \varepsilon_i)\Theta(\varepsilon_F - \varepsilon_i) \tag{13}$$

where N_i is the number of electrons per unit area in the ith subband. The self-consistent solution of (7)–(13) gives us the charge-density profile, the Fermi energy, the subband energies, and the total potential.

Since $\alpha \ll 1$ (i.e., $\alpha = 0.06$ in GaAs), we assume the electron to be almost free in the transverse directions and thus utilize a plane-wave representation for its motion parallel to the x–y plane, i.e., we take $\phi_k(\rho) \sim \exp(i\boldsymbol{k}\cdot\rho)$. We begin by expressing the general total wave function in a product form of the electron and phonon parts, i.e., $\psi = \Phi_{k,n}(\rho, z)\chi_{ph} = \varphi_n(z)\phi_k(\rho)\chi_{ph}$. For the ground state we take the electron to be in the lowest subband ($n = 0$) and select χ_{ph} as the phonon vacuum $|0\rangle$ simply because at low temperatures ($kT \ll \hbar\omega_{LO}$) there will be no effective phonons.

Since in the most commonly studied compound materials (such as GaAs) the electron–phonon coupling is rather weak, an appropriate approach is to treat the Fröhlich interaction H_{e-ph} as a perturbation. Up to first order,

$$|\Psi_{k,m,ph}\rangle = |0\rangle|\Phi_{k,m}\rangle + \sum_Q \sum_{k'} \sum_n \frac{\langle\Phi_{k',n}|\langle 1_Q|H_{e-ph}|0\rangle|\Phi_{k,0}\rangle}{\varepsilon_n(k') - \varepsilon_0(k) + 1}|1_Q\rangle|\Phi_{k',n}\rangle. \tag{14}$$

Now we will present a formalism for the derivation of optical rectification in this model. Let us consider an electromagnetic field of frequency ω which is incident with a polarization vector normal to the quantum wells. The system is excited by an internal electromagnetic field $E(t) = E_0 \cos\omega t$. The electronic polarization of the system due to the internal field $E(t)$ can be expressed as

$$\boldsymbol{P}(t) = \varepsilon_0\chi^{(1)}(\omega)\boldsymbol{E}e^{-i\omega t} + \varepsilon_0\chi^{(2)}_{2\omega}(\omega)\boldsymbol{E}^2e^{-2i\omega t} + \text{CC} + \varepsilon_0\chi^{(2)}_0(\omega)\boldsymbol{E}^2 \tag{15}$$

where $\chi^{(1)}(\omega)$ and $\chi^{(2)}_{2\omega}(\omega)$ are the linear and the second-order optical susceptibility coefficients. The last term of (15) indeed predicts the generation of a dc electric field by an optical beam; $\chi^{(2)}_0$ is the optical rectification coefficient. Using the same density matrix formalism as in [12] and [13], we have calculated the expression of $\chi^{(2)}_0$ for this model:

$$\chi^{(2)}_0 = 2\frac{e^3\sigma_s}{\varepsilon_0\hbar^2}\mu^2_{01}\delta_{01}\frac{\omega^2_{10}}{[(\omega_{10}-\omega)^2+\gamma^2][(\omega_{10}+\omega)^2+\gamma^2]} \tag{16}$$

where $\mu_{01} = |\langle\Psi_{k,0,ph}|z|\Psi_{k,1,ph}\rangle|$, $\delta_{01} = \langle\Psi_{k,1,ph}|z|\Psi_{k,1,ph}\rangle - \langle\Psi_{k,0,ph}|z|\Psi_{k,0,ph}\rangle$, e is the electronic charge, $\omega_{10} = (\varepsilon_1 - \varepsilon_0)/\hbar$ is the Bohr frequency, $1/\gamma$ is the dephasing time of the ground state and first excited state, and σ_s is the density of electrons in the parabolic quantum well. The optical rectification coefficient is at its maximum near resonance ($\omega \sim \omega_{10}$, $\gamma \ll \omega_{10}$):

$$\chi^{(2)}_{0,\max} = \frac{e^3\sigma_s}{2\varepsilon_0\hbar^2\gamma^2}\mu^2_{01}\delta_{01}. \tag{17}$$

3. Results and discussions

For a given concentration of carriers, the optical rectification coefficient may be optimized by choosing a structure maximizing the product of the oscillator strength with the induced dipole (proportional to $\mu_{01}^2\delta_{01}$). In our model, we can tune independently the force constant D in the parabolic potential and the applied electric field to optimize $\mu_{01}^2\delta_{01}$.

As an illustration, $\mu_{01}^2\delta_{01}$ is computed as a function of the electric field F for three different force constants D, and the results are shown in figure 1. From figure 1, we show that $\mu_{01}^2\delta_{01}$ increases with the enhancement of the applied electric field F. As the force constant D increases, $\mu_{01}^2\delta_{01}$ decreases. We note that the magnitude of $\mu_{01}^2\delta_{01}$ is proportional to the applied bias field F.

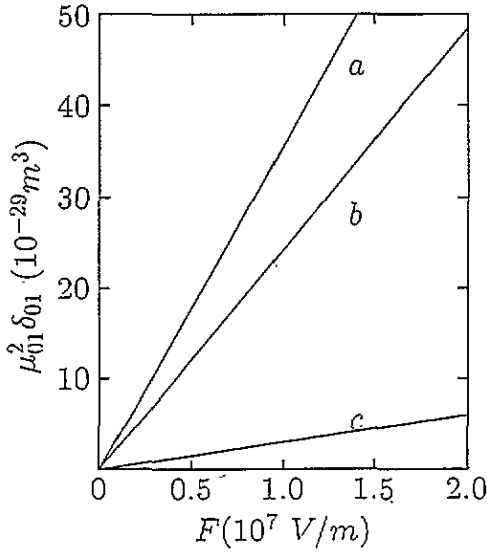

Figure 1. Variation of the product of dipolar matrix elements $\mu_{01}^2\delta_{01}$ as a function of the electric field F. It is plotted for three different force constants D in the parabolic potential: (*a*) $D = 5.0 \times 10^{-8}$ kg m s^{-2}; (*b*) $D = 1.0 \times 10^{-7}$ kg m s^{-2}; (*c*) $D = 1.5 \times 10^{-7}$ kg m s^{-2}. It shows that $\mu_{01}^2\delta_{01}$ increases with the enhancement of the applied electric field F, and increases with the decrease of the force constant D in the parabolic potential.

In figure 2, the optical rectification $\chi_0^{(2)}$ is plotted as a function of the energy of the fundamental photon. The best set of parameters we have found for optimum optical rectification are $\sigma_s = 5 \times 10^{24}$ m^{-3}, $F = 2.0 \times 10^7$ V m^{-1}, and $D = 50$. We have also used the usually quoted value for $1/\gamma$ of 0.14 ps [14]. Our curve is compared with the one given in [10], in which the electron–LO-phonon interaction is ignored. We can see that the value of $\chi_0^{(2)}$ of this study is over 10 times larger than that of [10]. This means that, when we take account of the electron–LO-phonon interaction, the theoretical value of $\chi_0^{(2)}$ will be more pronounced. It is interesting that both in figure 1 and in figure 2, the results are independent of the electron–electron interaction and of the number of electrons in the parabolic quantum well.

Next we will give the physical explanations for this polaron enhancement. First, it is known that the LO phonon in ionic crystals involves the relative motion of positive and

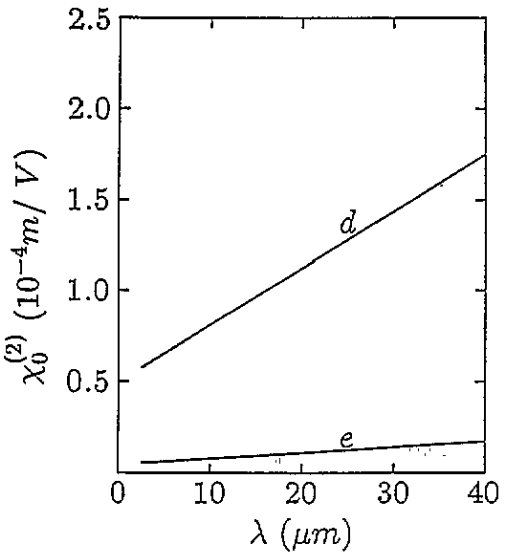

Figure 2. Calculated optical rectification $\chi_0^{(2)}$ as a function of the energy of the fundamental photon. The results are compared with those in [10], (*d*) taking account of the electron–LO-phonon interaction (this study); (*e*) ignoring the electron–LO-phonon interaction [10].

negative ions. This follows polarization and has a strong interaction with electromagnetic waves. Thus the LO phonon has a marked influence on the optical properties of ionic crystals. Second, in the literature, the so-called optical rectification usually refers to the generation of a dc electric polarization by an intense monochrome laser beam in a nonlinear medium. This dc electric polarization does not radiate electromagnetic waves, but forms a dc electric field in the medium. Therefore, when we take account of the electron–LO-phonon interaction, the dc electric polarization induced by the laser beam will increase notably. Thus, it is not surprising that the optical rectification coefficient scales by as much as a factor of 10, while the electron–LO-phonon interaction is taken into account. For the above reasons, the result of the paper should be of importance to future calculations of the second-order susceptibility in low-dimensional structures, and thus the work is useful and should be of relevance to experiment.

In conclusion, we have studied the optical rectification of the weak-coupling optical polaron systems in electric-field-biased parabolic quantum well media. Of particular interest is the electron–LO-phonon interaction and the fact that the results are independent of the electron–electron interaction and of the number of the electrons in the parabolic quantum well. Because of taking account of the electron–LO-phonon interaction, the theoretical value of $\chi_0^{(2)}$ is in good agreement with the experimental measurements. The results open up the way to synthetic nonlinear semiconductors for future use in far-infrared signal processing.

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