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Effect of a static electric field on the optical spectrum of a quantum dot

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Abstract

The energy levels and the optical absorption of a circular quantum dot with hard walls are studied in the presence of a static electric field. The results are compared with the case of soft-wall confinement. Calculations show that: (i) the structures of the energy levels of the two cases are different, therefore their absorption varies accordingly; (ii) the nonlinear term has a significant contribution to the total optical absorption coefficient. © Elsevier Science B.V.

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1. Introduction

In recent years, quantum dots have become one of the foci of interest in nanometer science [1–5]. It is a kind of fabricated semiconductor device in which the electrons are confined in all spatial directions and therefore a quasi-zero-dimensional (Q0D) electron gas is formed. Two theoretical models are widely used to describe quantum dots. One of them is called soft-wall model, in which a parabolic confining potential is chosen [6–8]. Such a potential always leads to exact solutions of the energy levels. This model is probably suitable for representing the quantum dots provided by state-of-the art nanolithography techniques, where the quantum dots have been fabricated by electrostatic confinement of a two-dimensional electron gas. Based on etching techniques which are able to fabricate columns that contain a Q0D electron gas, an alternative technique is expected to provide more well-defined confinement walls, and in the extreme limit these kinds of quantum dots may be well represented by the hard-wall model, in which the confining potential takes the form of an infinite or finite square well [9–12]. Usually the calculations involved in this kind of model have to be carried out numerically.

The optical absorption of quantum wells has become the subject of many studies lately. Ahn and Chuang [13] have derived the linear and nonlinear optical absorption spectra taking the density matrix formalism with intra-subband carrier relaxations into account, Kuhn et al. [14] have adopted the finite difference method to

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calculate the energy levels and the change in the real part of the dielectric constant, and they all found that the contribution of the nonlinear term to the total absorption coefficient is quite significant. Recently Panda et al. [15] have calculated the effects of the electric field on the electron subband energies and wavefunctions both in a single infinite quantum well and in a finite one. So the detailed knowledge of the optical absorption spectra in quantum dots should also be of interest for device applications.

Electric and magnetic fields are both important methods to study the optical spectrum of semiconductors. Geerinckx et al. [16] have calculated the magneto-optical spectrum of a quantum dot using both models and results showed that in contrast to the parabolic case where only two transition energies are found, in the hard-wall case there are many transitions possible which have different energies. However, only a small number of them have sufficient oscillator strength to be observable. In this paper, we calculate the effect of a static electric field on the energy levels of a two-dimensional quantum dot described by both the soft-wall and hard-wall models, and take a look into their optical absorption properties.

2. Soft-wall model and exact solution

In this case the confining potential of the quantum dot is taken to be in a quadratic form, that is

$$V(r)=\tfrac{1}{2}m^*\omega_0^2r^2, \tag{1}$$

where $r=\sqrt{x^2+y^2}$ for a two-dimensional system, m^* is the effective electron mass, and ω_0 is the effective oscillator frequency. We apply a static electric field F along the x direction, and the single-particle Hamiltonian for an electron reads $H=H_x+H_y$, with

$$H_x=\frac{p_x^2}{2m^*}+\tfrac{1}{2}m^*\omega_0^2x^2+eFx,\qquad H_y=\frac{P_y^2}{2m^*}+\tfrac{1}{2}m^*\omega_0^2y^2. \tag{2}$$

Here the effect of spin is neglected. Then the Schrödinger equation $H\psi(x,y)=E\psi(x,y)$ gives both $H_x\psi(x)=E_x\psi(x)$ and $H_y\psi(y)=E_y\psi(y)$, with $\psi(x,y)=\psi(x)\psi(y)$, $E_x+E_y=E$. An exact solution can easily be abtained, and the result is

$$\psi(x)=N_x\,\mathrm{e}^{-\alpha^2x'^2/2}H_{n_x}(\alpha x'),\qquad E_x=\left(n_x+\tfrac{1}{2}\right)\hbar\omega_0-\frac{e^2F^2}{2m^*\omega_0^2},$$

and

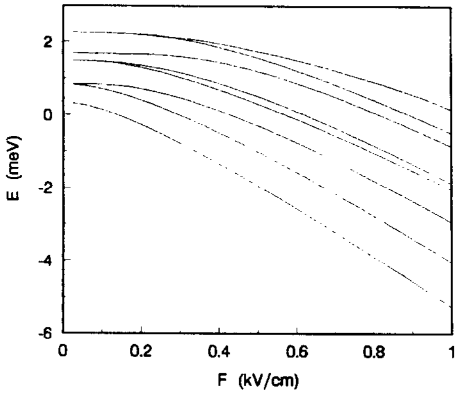
$$\psi(y)=N_y\,\mathrm{e}^{-a^2y^2/2}H_{n_y}(\alpha y),\qquad E_y=\left(n_y+\tfrac{1}{2}\right)\hbar\omega_0, \tag{3}$$

where $\alpha\sqrt{m^*\omega_0/\hbar}$, $x'=x+eF/m^*\omega_0^2$, N_x and N_y are normalization constants, n_x and n_y are quantum numbers, and H_n is the n-degree Hermite function.

Then from the expression of the total energy

$$E=E_x+E_y=\left(n_x+n_y+1\right)\hbar\omega_0-\frac{e^2F^2}{2m^*\omega_0^2} \tag{4}$$

we can find an important conclusion: changing the magnitude of the applied electric field will only produce a shift to every energy level of the system, while the space between all energy levels will remain unchanged, so the frequency of the photons emitted when the system jumps between energy levels will not be affected by the



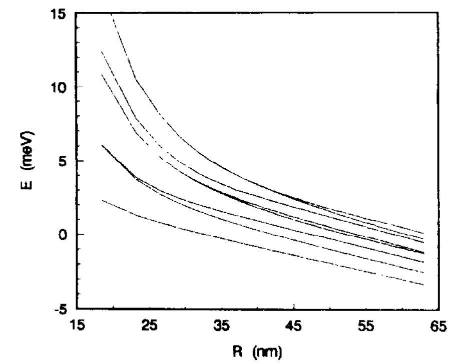


Fig. 1. The energies of the first eight energy levels as a function of the intensity of the electric field with $R = 50$ nm.

Fig. 2. The energies of the first eight energy levels as a function of the radius of the quantum dot with $F = 50$ kV/cm.

electric field. In the following we shall see that this conclusion is derived from the specific model used here to describe quantum dots, and will not be valid in the hard-wall model.

3. Hard-wall model and numerical result

An infinite hard-wall model contains a confining potential with the following form,

$$V(r) \begin{array}{ll} = 0, & r \leqslant R, \\ = +\infty, & r > R, \end{array} \tag{5}$$

where R is the radius of the quantum dot. Then the Hamiltonian for the same problem becomes

$$H = \frac{p_x^2 + p_y^2}{2m^*} + eFx + V(r), \tag{6}$$

with the boundary condition

$$\psi(x, y)\,|_{x^2+y^2=R^2} = 0. \tag{7}$$

No exact solution can be obtained for the Schrödinger equation $H\psi(x, y) = E\psi(x, y)$ in this case, and we have to solve it numerically. The result is shown in the following figures.

Fig. 1 shows the eigenenergies of the system as a function of the intensity of the electric field with the radius R of the quantum dot fixed at 50 nm, while Fig. 2 shows the relationship between energies and quantum dot radius R with electric field F fixed at 50 kV/cm. From these figures we can easily see that the energy levels drop as the electric field or the size of the quantum dot increases, and this tendency agrees with the soft-wall model (see Eq. (4), where the decrease of ω_0 is equivalent to the increase of R), but under the current condition the degenerate energy levels are split into doublets as the electric field increases, and the larger the quantum dot is, the clearer this effect is. So this time the spaces between the energy levels do change as the electric field varies. We made this clearer in Fig. 3, which shows the difference in energy of the first four energy levels with R fixed at 50 nm. In the following we will study the influence of this difference in energy level structure on the optical absorption properties of the two QD models.

4. Optical absorption and change in the real part of the refractive index

If the quantum dot is subjected to an optical intensity, there will be a dc rectification term and second- and higher-harmonic generation terms [17] in addition to the linear and nonlinear polarization terms. Keeping the linear and nonlinear susceptibilities only, we can obtain

$$P(t) = \varepsilon_0 \chi^{(1)}(\omega) I_{\mathrm{op}}\,\mathrm{e}^{-\mathrm{i}\omega t} + \varepsilon_0 \chi^{(3)}(\omega) I_{\mathrm{op}}\,\mathrm{e}^{-\mathrm{i}\omega t} + \varepsilon_0 \chi^{(1)}(\omega) I_{\mathrm{op}}^{*}\,\mathrm{e}^{\mathrm{i}\omega t} + \varepsilon_0 \chi^{(3)}(\omega) I_{\mathrm{op}}^{*}\,\mathrm{e}^{\mathrm{i}\omega t}, \tag{8}$$

where I_{op} is the amplitude of the optical source. The expression for the linear susceptibility $\chi^{(1)}(\omega)$ is given by [13,14]

$$\varepsilon_0 \chi^{(1)}(\omega) = |M_{\mathrm{fi}}|^2 \left(\frac{m^* k_B T}{L\pi\hbar^2} \right) \ln\left(\frac{1+\exp[(E_F - E_i)/k_B T]}{1+\exp[(E_F - E_f)/k_B T]} \right) \frac{1}{E_f - E_i - \hbar\omega - \mathrm{i}\hbar/\tau_{\mathrm{if}}}, \tag{9}$$

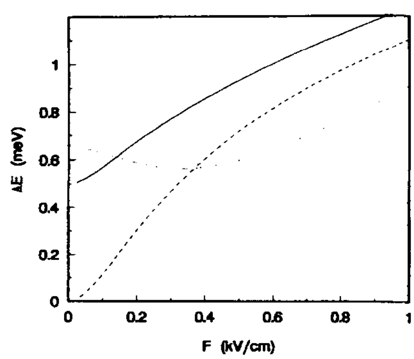
where E_F is the Fermi energy, which is taken to be 0.023 eV [15]. E_i and E_f are the initial and final energy respectively. m^* is the effective mass in the quantum dot. $L = 2R$ is the effective width of the QD. M_{ab} is the dipole matrix element between states $|a\rangle$ and $|b\rangle$ described as

$$M_{ab} = |e| \int \psi_a(z)\, z \psi_b(z)\, \mathrm{d}z, \tag{10}$$

and τ_{ab} is the dephasing rate, which is the combination of the rates from the electron tunneling time, electron–phonon scattering, ionized impurity scattering and interface roughness scattering, and it is given by

$$\frac{1}{\tau_{ab}} = \frac{1}{2}\left(\frac{1}{\tau_a} + \frac{1}{\tau_b} \right). \tag{11}$$

In the following calculations we assumed τ_{ab} to be a constant and to have the experimental value of 0.14 ps when $T = 77$ K [13].



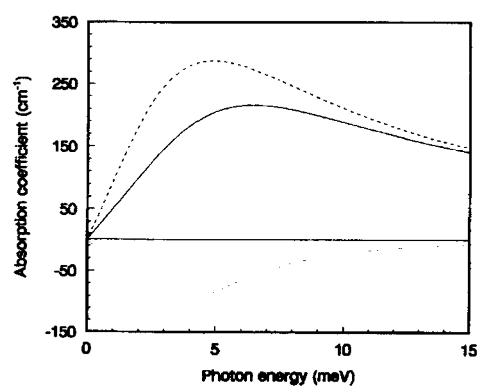


Fig. 3. The energy difference between two energy levels as a function of the intensity of the electric field with $R = 50$ nm. The three curves are for the energy difference between: the first and second energy levels (solid line); the second and third energy levels (dashed line); the third and fourth energy levels (dotted line).

Fig. 4. Linear (dashed line), nonlinear (dotted line), and total (solid line) absorption coefficients for the soft-wall model for a zero applied dc field. The initial and final energy levels are the first two energy levels of the system. The incident optical intensity is 0.01 MW/cm^2.

The third-order nonlinear susceptibility is given by

$$\varepsilon_0 \chi^{(3)}(\omega, I) = -\varepsilon_0 \chi^{(1)}(\omega)\left(\frac{1}{2\varepsilon_0 n_r c}\right)\left(\frac{2(\tau_{ii}+\tau_{ff})\,|M_{fi}|^2}{\tau_{if}\left((E_f-E_i-\hbar\omega)^2+\hbar^2/\tau_{if}^2\right)} - \frac{(M_{ff}-M_{ii})^2}{(E_f-E_i-i\hbar/\tau_{if})(E_f-E_i-\hbar\omega-i\hbar/\tau_{if})}\right), \tag{12}$$

where $I = |I_{op}|^2$ is the intensity of the optical source. c is the speed of light in free space. Then the absorption coefficient $\alpha(\omega, I)$ is obtained

$$\alpha(\omega, I) = \omega\sqrt{\frac{\mu}{\varepsilon_R}}\,\mathrm{Im}[\varepsilon_0 \chi(\omega)] \tag{13}$$

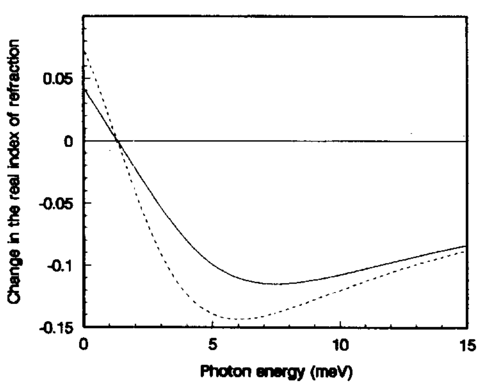
and the change in the refractive index is related to the susceptibility as

$$\frac{\Delta n(\omega)}{n_r} = \mathrm{Re}\left(\frac{\chi(\omega)}{2n_r^2}\right) \tag{14}$$

where ε_R ($\varepsilon_R = n_r^2\varepsilon_0$) is the real part of the permittivity, n_r is the refractive index, whose value is 3.2 for GaAs, and μ is the permeability of the material.

5. Result and discussion

In order to make a comparison between the two models, we take the value of the parameter ω_0 in the soft-wall potential to be 1.988×10^{12} s^{-1}, which gives the same ground state energy value as that of a hard-wall model with a radius of 50 nm when no external electric field is applied. Taking only the ground state and the first excited state into consideration, the absorption coefficient and the change in the real index of the refraction for the soft-wall model for a zero applied dc field are shown in Figs. 4 and 5. Comparing with the results for the quantum well obtained by Panda et al. [15], we can see that the contribution of the nonlinear term



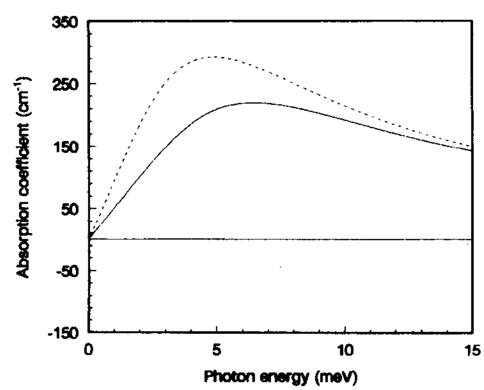
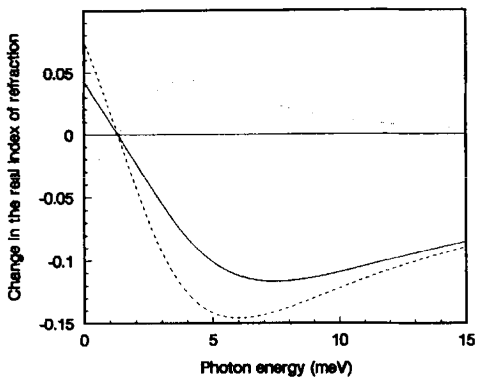


Fig. 5. Linear, nonlinear, and total change in the real part of the index of refraction for the soft-wall model for a zero applied dc field. The parameters and the notations are the same as in Fig. 4.

Fig. 6. Linear, nonlinear, and total absorption coefficients for the soft-wall model for a 1 kV/cm applied dc field. The parameters and the notations are the same as in Fig. 4.



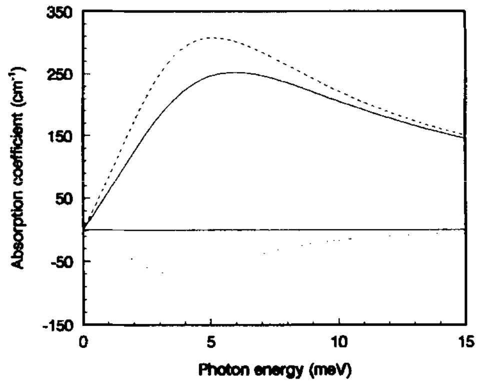


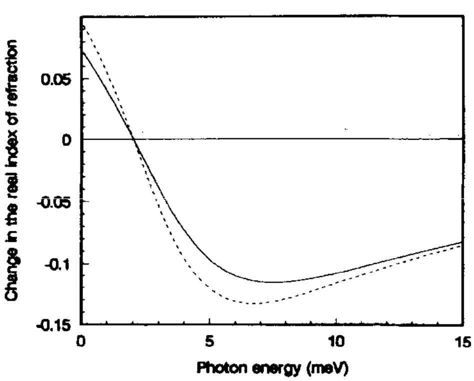
Fig. 7. Linear, nonlinear, and total change in the real part of the index of refraction for the soft-wall model for a 1 kV/cm applied dc field. The parameters and the notations are the same as in Fig. 4.

Fig. 8. Linear, nonlinear, and total absorption coefficients for the hard-wall model for a zero applied dc field. The parameters and the notations are the same as in Fig. 4.

to the total absorption coefficient is opposite to that of the linear term in both cases, but the nonlinear term in the quantum dot is much larger (about 100 times) in quantity (noting that the incident optical intensity used in Ref. [15] is 1 MW/cm^2). This may be due to the size of the QD, which is about one order of magnitude smaller than that of the QW. Further calculations show that the ratio between the linear and nonlinear terms in the QD does tend to the QW value as we decrease the radius of the QD.

Figs. 6 and 7 show the results of the same system studied in Figs. 4 and 5, except that a 1 kV/cm static electric field is applied. Since the degenerate energy levels in a soft-wall model do not split with the increased electric field, it is not surprising that the absorption coefficient and the change in the real index of refraction change only little.

For a hard-wall model with $R = 50$ nm, the absorption coefficient and the change in the real index of refraction caused by the transition between the first two energy levels show the same property as that of the soft-wall potential when the electric field vanishes (see Figs. 8 and 9). But when the field is applied, both the absorption coefficient and the change in the real index of refraction drop to such an extent that they are actually



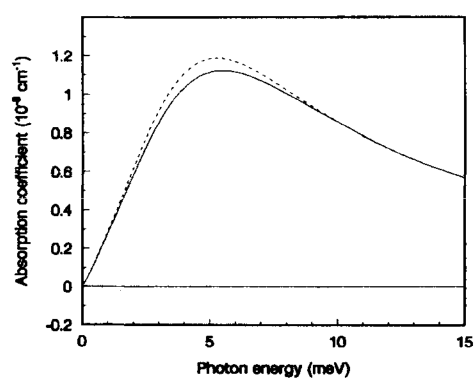
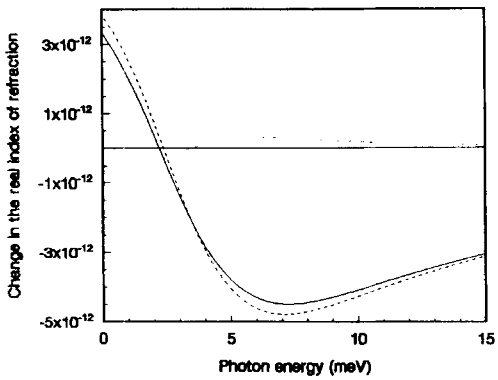


Fig. 9. Linear, nonlinear, and total change in the real part of the index of refraction for the hard-wall model for a zero applied dc field. The parameters and the notations are the same as in Fig. 4.

Fig. 10. Linear, nonlinear, and total absorption coefficients for the hard-wall model for a 1 kV/cm applied dc field. The notations are the same as in Fig. 4 except that the incident optical intensity is 0.1 MW/cm^2.



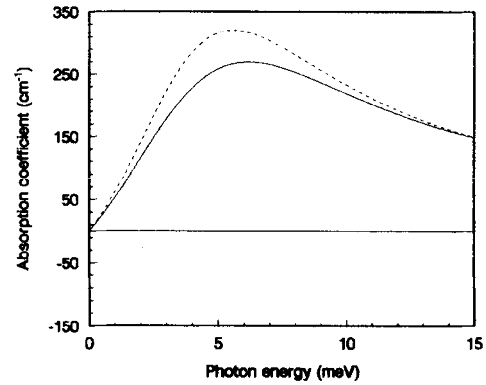
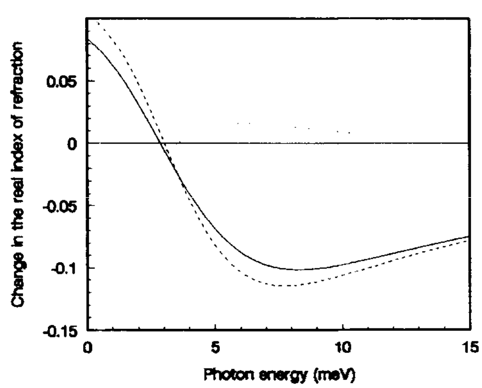


Fig. 11. Linear, nonlinear, and total change in the real part of the index of refraction for the hard-wall model for a 1 kV/cm applied dc field. The parameters and notations are the same as in Fig. 10.

Fig. 12. Linear, nonlinear, and total absorption coefficients for the hard-wall model for a 1 kV/cm applied dc field. The initial and final energy levels are the first and the third energy levels of the system. Other parameters and notations are the same as in Fig. 4.

negligible compared with the zero field values, as it is shown in Figs. 10 and 11. To understand this phenomenon, we studied the transition between the first and the third energy levels, and the results are shown in Figs. 12 and 13.We can see now that the optical parameters have turned back to the magnitude in the zero field case. Since the second and the third energy levels originate from the same degenerate energy level, we can see that though the increasing electric field splits the original single level into doublets, the split of the absorption spectrum related to the transitions between the ground state and these energy levels is not observable. This is quite similar to the magneto-optical spectrum obtained by Geerinckx et al. [16].

The transitions involving higher excited states are shown in Fig. 14. We find that the function of the external static electric field is splitting the entire energy levels into two sets, the transitions inside each set have larger dipole matrix elements, while the transitions between the two sets are much less observable.



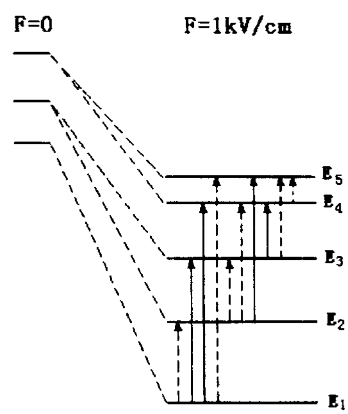


Fig. 13. Linear, nonlinear, and total change in the real part of the index of refraction for the hard-wall model for a 1 kV/cm applied dc field. The parameters and notations are the same as in Fig. 12.

Fig. 14. Transitions between the first five energy levels for the hard-wall model for a 1 kV/cm applied dc field. Transitions depicted by dashed lines have dipole matrix elements that are at least three orders of magnitude smaller than those depicted by solid lines.

6. Conclusion

From the above we can see that the nonlinear optical properties in a quantum dot are much more distinct than those in a quantum well, and since a static electric field will not split the degenerate energy levels in a soft-wall potential as it does in the hard-wall model, the optical absorption spectrum in the latter is more complex. It is therefore necessary to pay more attention to the nonlinear optical properties of QD.

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