Ground state transition energies in biased InAs/GaAs quantum dots

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Abstract

In this work, we report on the calculation of the electron–hole ground-state transition energies in InAs/GaAs quantum dots. We examine how the external electric field and the basis radius $r_c$ of the quantum dots affect the electron–hole ground-state transition energies. The results presented in this work are in sound agreement with recent experimental observations.

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1. Introduction

The rapid progress in semiconductor technology in recent years has allowed the fabrication of low-dimensional electronic nanostructures, quantum dots (QDs), wires, etc. These nanostructures are characterized by a lateral confinement of a quasi-two-dimensional electron gas leading to a discrete energy spectrum observable in low temperature experiments [1].

In recent years, InAs/GaAs self-assembled QDs have been the subject of intense interest as a result of their zero-dimensional electronic states [2], their excellent optoelectronic quality, and their considerable contemporary interest for applications such as ultra low threshold lasers [2] and memory devices [3]. Despite their favourable applications, many questions regarding their electronic properties remain unknown. In particular, less experimental information is available about the nature of QDs carrier wave functions and their response to an applied electric field [4–6]. Moreover, it is now experimentally well established that nanometer QDs can be fabricated by self-organized...
growth in strongly lattice-mismatched systems, for instance InAs dots embedded in a GaAs matrix [4–14]. These InAs dots emit in the 1 eV range depending on their basis radius $r_c$.

The paper is organized in the following way. In Section 2, we present the model and our numerical approach. In Section 3, the electron and hole energy levels have been calculated. This is followed in Section 4 by the presentation of how the basis radius of the QDs and applied electric field affect the electron–hole ground-state transition energies. Finally, in Section 5 we summarize the main conclusions.

2. Model

In this work, we are interested in the numerical description of the electron–hole ground-state transition energies in InAs/GaAs QDs. As shown by atomic force microscopy and deduced from the optical study of InAs quantum wells in GaAs [12,15], QDs consist of small truncated pyramids with a circular base of radius $r_c$. In our calculations, QDs are modelled by cones (height $h$ and radius $r_c$) as shown in Fig. 1. These pyramids have the same height $h$ in the $z$ growth direction and the same radius $r_c$ in the $\rho$ direction. The base angle of the cone is close to $30^\circ$ for $h = 3 \text{ nm}$.

We have calculated, independently, the electron and heavy hole energy levels, neglecting light hole effect, excitonic effects and spin–orbit effect. Due to cylindrical symmetry of the system, the problem is 2D and the quantum dot confinement potential $V_{\text{conf}}$ couples the motion along growth direction $z$ and radial coordinate $\rho$. The noninteracting Hamiltonian describing a single particle (electron or hole), in the effective mass approximation is given by

$$H_0 = T_\rho + V_{\text{conf}} + V_F,$$

where $T_\rho$ denotes the kinetic energy operator and $V_F$ is the electrostatic potential introducing the effects of static electric field applied parallel to the cone axis $z$. In the cylindrical symmetry, the three terms of the Hamiltonian are given by

$$T_\rho = \left(-\frac{\hbar^2}{2m^*_\rho}\frac{\partial}{\partial \rho} \left( \rho \frac{\partial}{\partial \rho} \right) + \frac{1}{\rho^2} \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial z^2} \right),$$

$$V_F = -q_0Fz_c,$$

$$V_{\text{conf}}(\rho, z_c) = V_c\Theta \left[ z_c - \left( (r_c - \rho) \tan(z) + \frac{Z+d-h}{2} \right) \right] \times \Theta \left[ \frac{Z+d+h}{2} - z_c \right] \Theta[r_c - r_c] \Theta[r_c - \rho_c] $$

$$+ V_c\Theta \left[ z_c - \frac{Z+d-h}{2} \right] \Theta \left[ \frac{Z+d+h}{2} - z_c \right] \times \Theta[r_c - r_c] \Theta[R - \rho_c] $$

$$+ V_c\Theta \left[ z_c - \frac{Z+d-h}{2} \right] \times \Theta[Z - z_c] \Theta[\rho_c] \Theta[R - \rho_c] $$

$$+ V_c\Theta \left[ \frac{Z - (d+h)}{2} - z_c \right] \times \Theta[z_c] \Theta[\rho_c] \Theta[R - \rho_c] $$.  \hfill (2.4)

where $\Theta$ is the Heavyside function, $V_c = 0.697 \text{ eV}$ and $V_h = 0.288 \text{ eV}$ are, respectively, the electron and hole potentials [16]. The electrostatic term in the Hamiltonian is evaluated over a large cylinder of radius $R = 40 \text{ nm}$ and height $Z = 16 \text{ nm}$ in which the truncated cone is placed. $F$ denotes the electric field applied in the growth direction ($z$) and $q$ is the particle charge, $d = 0.566 \text{ nm}$ is the thickness of the wetting layer and $h = 3 \text{ nm}$ corresponds to the truncated cone height. These numerical parameters used in our calculations correspond to an InAs/GaAs truncated pyramidal QDs at the temperature $T = 4.2 \text{ K}$ [16]. A Hamiltonian similar to Eq. (2.1) is considered for the holes in which the subscripts $h$ replace $e$. The electron and hole energy levels have been

![Fig. 1. Morphology of the QD observed in the z growth direction.](image)
numerically calculated by diagonalizing $H_0$ over the eigenstates basic of the large cylinder, given by

$$\Phi^m_n(\rho, z) = \alpha_i^n J_n(\frac{\rho}{R}) \sin(\pi j \frac{z}{Z}),$$

where $n$ denotes the main quantum number corresponding to S, P, D, F,... symmetries for the given values $n = 0, \pm1, \pm2, \pm3,...$ respectively. $\alpha_i^n$ is a normalization coefficient given by

$$\alpha_i^n = \sqrt{\frac{2}{\pi R^2}} \frac{1}{J_{n+1}(\lambda_i^n)},$$

$J_n$ is the $n$th Bessel function of integer order $n$ and $\lambda_i^n$ is the $i$th zero of $J_n$. In order to ensure a good convergence ($10^{-4}$ eV) of the calculated eigenvalues, we shall deal with a $20(i) \times 45(j)$ basis. The considered wave functions are displayed on the basis of the eigenfunctions of the Schrödinger equation of a free particle in the large cylinder. The truncated pyramidal quantum dot is placed on the axis of the large cylinder (see Fig. 1). The potential is taken to be infinite outside the cylinder, zero inside the InAs quantum dot and $V_{e,h}$ in the intermediate region between the cone and the large cylinder. In order to simplify the matter as much as possible, we make the assumption that the effective masses for the electron ($m_{e0} = m_{ez} = 0.067 m_0$) and for the hole ($m_{h0} = 0.11 m_0$ and $m_{hz} = 0.34 m_0$) are constant throughout, where $m_0$ is the free electron mass [16–18].

### 3. Energy levels

In this section, we are interested in the energy levels of the electron and hole for S, P, D and F symmetries. We study the radius $r_c$ and external electric field effects on these energy levels. The used numerical method enables us to calculate all energy levels, but we show only the first three levels for each symmetry. We report in Fig. 2 the energy levels for S, P and D symmetries versus $r_c$ for the electron and hole. Note that the electron and/or hole are in bound states when their energies are smaller than the wetting layer one. In fact, the wetting layer energies for the electron and hole are smaller than the ones of S symmetry corresponding to 667 meV for the electron and 258 meV for the hole (fundamental state energies for S symmetry), respectively. The states corresponding to the last energies perform the lower quasi-bound states. Sometimes, if this condition does not seem to be sufficient to know if the particle is inside or outside the QDs, we calculate the average of the cylindrical coordinate $z_0 = \langle \phi | z | \phi \rangle$ for each state $| \phi \rangle$ ($z_0$ is associated to the position along the $z$-axis of the particle, calculated using matrix method).

Note that when the particle (electron or/and hole) is in a bound state inside the truncated pyramidal QDs the average of its cylindrical coordinate $z$ verifies the following condition:

$$\frac{Z - (d + h)}{2} < z_0 < \frac{Z + (d + h)}{2}.$$  \hspace{1cm} (3.1)

For the considered QDs, the particle is in a bound state when the following condition is fulfilled:

$$6.217 \text{ nm} < z_0 < 9.783 \text{ nm}.$$  \hspace{1cm} (3.2)

We show, in Fig. 2, that the electron and hole are more and more bound in the QDs when the radius $r_c$ increases. Indeed, for $6 \text{ nm} < r_c < 8 \text{ nm}$ the S and P fundamental states of the electron and hole are bound in the QDs, whereas this is not the case for D symmetry particles since their energies are higher than that of the wetting layers. We also show that beyond $r_c = 8 \text{ nm}$, the fundamental states of the electron and hole for S, P and D symmetries are bound in the QDs. In order to make sure that the carriers are inside the QDs, we have calculated $z_0$ for each case. In particular for D symmetry, our calculations show that the electron and hole are in bound states only beyond $r_c = 8 \text{ nm}$. We have examined, at zero field, how $r_c$ affect $z_0$ for the electron and hole. Results are plotted in Fig. 3 for S and P symmetries. For these two symmetries, it is clear that the electron and hole are in bound states inside the truncated pyramidal QDs. Moreover, by varying $r_c$ from 6 to 10 nm, the electron is still for ever, at the quantum dot apex, whereas the hole is localized at the bottom. We remark, for each symmetry, that $z_0$ increases as a function of $r_c$ for the two carriers indicating that the electron and hole are more and more localized at the proximity of the QDs middle.

Fig. 4 depicts energy levels of the electron and hole as a function of the applied electric field for
$r_c = 8\text{ nm}$, only the first three levels for S and P symmetries are reported. Fig. 4 shows a linear variation of the electron energy levels, this is the linear Stark effect. From Fig. 4, we distinguish three regions exhibiting different behaviours of the hole energy levels. At the first region, we find the bound states where we have the linear Stark effect. The second kind of behaviour, characterized by deviation from the linear behaviour, is exhibited by the levels which belong to the quasi-bidimensional continuum at zero field. Such levels, at small fields, behave like truly bound states of the QDs and they are quickly admixed with a quasi-continuum of states originating from levels which are totally unbound in the pyramidal cone at zero electric field. These levels are strongly affected by the electric field. For high fields, we find that the lower energy levels are not localized in the dot, as at zero fields, but rather accumulate near one end of the large cylinder. According to experimental observations, this is due to the tunnel effect in the triangular part of the quantum dot [4–6].

The electric field dependence of $z_0$, for S and P symmetries, is plotted in Fig. 5. We show that at zero fields, for both the mentioned symmetries, the hole is localized towards the bottom of the quantum dot, below the electron. This result is in agreement with previous theories [19,20], with the hole wave function below that of the electron. On the other hand, it is important to note that our calculations of the relative position of the carriers in the QDs at zero electric field is in agreement with recent experimental observations made by Fry et al. [4] showing that QDs possess a permanent dipole moment, implying a spatial separation of the electron and hole (for $F = 0$),
Fig. 3. (a) Electron and hole $z_0$ for S symmetry versus $r_c$ at zero electric field. (b) Electron and hole $z_0$ for P symmetry versus $r_c$ at zero electric field.

Fig. 4. Electron (a) and hole (b) energy levels for S, P and D symmetries versus applied electric field for $r_c = 8$ nm. Only first three levels are reported for each symmetry: the fundamental state ($S^0$, $P^0$ and $D^0$), the first ($S^1$, $P^1$ and $D^1$) and the second ($S^2$, $P^2$ and $D^2$) excited states.
and then sensitive to the applied electric field. The dipole moment is expected from the asymmetric shape of the QDs. According to previous results, the applied electric field $F$ in the direction from the base to the apex of the QDs (i.e. in the growth direction) will lead to electron (hole) attraction to the base (apex) of the dots. These predictions are confirmed by our calculations of $z_0$ versus $F$ plotted in Fig. 5. In fact, we find reversal of this alignment taking place around an applied electric field of 50 kV/cm.

Recent experimental observations found that the electron is localized at the apex and the hole at the base at zero fields only if two conditions are fulfilled [4]. Firstly, the dots must have a graded $\text{In}_{1-x}\text{Ga}_x\text{As}$ composition, where $x$ denotes the gallium concentration and decreasing from base to apex. So, it was shown that the holes tend to be localized in the region with the largest indium concentration. Secondly, it was also necessary to severely truncate the pyramid to achieve a dipole pointing to the QDs base, since strain localizes the hole strongly below the electron until the truncation factor is greater than 0.6 [22]. For our calculations, the truncation factor corresponding to $h = 3$ nm is close to 0.65 corresponding to localized hole wave function below that of the electron. Our results are in good agreement with Fry et al.’s works [4].

It is worth pointing out that from our study of the electron and hole energy levels with respect to $r_c$ and the applied electric field, we distinguish several sorts of behaviours. At zero fields, the electron and hole bound levels decrease with increasing $r_c$ and other levels, which are totally unbound in the truncated cone, go down from the quasi-2D continuum when $r_c$ increases (see Fig. 2). The main remark that we can make is that the two first bound states are more than 50 meV apart. The electron and hole states are strongly localized in the QDs, their energies are 371.1 meV and 130.9 meV to be compared to $V_e$ and $V_h$, respectively. This strong localization implies rigidity of the bound states towards external perturbation, like coulombic effects and external electric field effect.
The calculated energy levels of the electron and hole versus $F$ for a given value of the basis radius ($r_c = 8$ nm), reported in Fig. 4, show a linear Stark effect. In fact, for $r_c = 8$ nm, when the applied electric field increases, the electron and hole are more confined. As shown in Fig. 4, the fundamental states corresponding to S and P symmetries are lying in the QDs until $F = 200$ kV/cm. For D symmetry, the fundamental electron level is bound in the QDs for $100$ kV/cm < $F < 180$ kV/cm. In fact, for $F < 100$ kV/cm, the electron energy for D symmetry is greater than the wetting layer one and for $F > 180$ kV/cm the calculated value of $z_0$ shows that the electron is outside the QDs.

It is essential to specify that we are interested in this part of this work to examine how basis radius and applied electric field affect energy levels of the electron and hole in InAs/GaAs QDs. So, we are interested precisely to carriers which are spontaneously inside the QDs or by varying $r_c$ or/and the electric field. Our study of the carriers behaviour in the QDs provides a plausible explanation of recent experimental observations [4–6]. In fact, Fry et al. employed photoluminescence spectroscopy and photocurrent spectroscopy on InAs/GaAs quantum dots (having $h \approx 3$ nm of height and $r_c \approx 8$ nm of basis radius, the same that we use for our modelled QDs). For above-gap excitation, inhibition of capture is found at very small electric fields. For carriers excited in the wetting layer, direct capture into the QDs dominates at low fields, but when the applied electric field increases, tunnel escape from the wetting layer dominates (we show that the calculated values of $z_0$ enable us to examine this case). For direct excitation in the QDs, it was shown the occupation of the active region is much more stable, with tunnel escape important only when the applied electric field exceeds 100 kV/cm. These observations are reinforced by photocurrent spectroscopy which is a direct and sensitive technique to measure low-noise absorption spectra of QDs [23]. Our calculations of the energy levels of the electron and hole and then the corresponding $z_0$, are in sound agreement with these experimental observations. On the other hand, our calculations assuming pure InAs QDs predict positive dipole moments, with the hole confined at the base of the dot, below the electron. This result is in agreement with previous calculations with the same assumption [19–21].

### 4. Ground state transition energies

In this section we are interested to the electron–hole ground-state transition energies given by

$$T = e_n^{e} + e_n^{h} + g,$$

(4.1)

where $n$ indicates the symmetry, $e_n^{e}$ and $e_n^{h}$ are the electron and hole energies at the fundamental states. Note that the gap energy $g$ is not computed when we plot the transition energies because its effect is simply a shift of all energies. It is important to note that at low temperature only the ground-state transition energies are detected. Thermal emission is important only for temperatures exceeding 150 K [4,24]. Our calculations of the ground-state transition energies are done according to these considerations. The numerical parameters used in the calculations are given for 4.2 K [16].

The transition energies are evaluated with respect to the basis radius $r_c$ of the QDs and the applied electric field $F$. For the considered symmetries, only the bound states in QDs are
considered. Evolution of the ground state transition energies as a function of \( r_c \) is presented in Fig. 6 for S, P and D symmetries. We note that these energies decrease for all symmetries when \( r_c \) increases. This behaviour is ascribed to particles confinement. In fact, we show in Fig. 7, for S and P symmetries, that the electron and hole are more and more bound closer when \( r_c \) increases. Fig. 8 displays the evolution of ground state transition energies as a function of \( F \) for both S and P symmetries. We remark a parabolic shape attributed to the electron and hole inversion phenomena, where the maximum corresponds to \( F_0 \) (the applied electric field necessary for the inversion). Our calculations of the field dependence of the ground state transition energies \( T \) is in good agreement with experimental results [4,5] and it can be described with a very reasonable approximation by the expression

\[
T = T_0 + pF + \beta F^2,
\]

where \( T_0 \) is the zero fields energy depending on \( h \), the second term \( (pF) \) arises from the nonzero dipole moment \( p \), and the third one arises from the quantum confined Stark effect leading to polariza-

![Fig. 7. \((\langle z_e^0 \rangle - \langle z_h^0 \rangle)\) for S and P symmetries plotted as a function of \( r_c \) at zero electric field.](image)

![Fig. 8. (a) Ground state transition energy of electron–hole for S symmetry reported as a function of applied electric field for \( r_c = 8 \text{ nm} \). (b) Ground state transition energy of electron–hole for P symmetry reported as a function of applied electric field for \( r_c = 8 \text{ nm} \).](image)
tion of the quantum dots in the applied electric field. The quadratic component $\beta$ of the energy shift is essentially related to the QDs height $h$ [4,25]. We can show easily that for each value of the QD height, $p$ and $\beta$ are related by

$$\beta = -\frac{p}{2F_0}.$$  \hfill (4.3)

We also show that the maximum ground state transition energy for S and P symmetries occurs for $F_0 \approx 50 \text{kV/cm}$ (when the inversion phenomena takes place). These results are coherent with evolution of $(z_e^0 - z_h^0)$ presented in Fig. 9 showing, for S and P symmetries, the quantum confined Stark effect. We have calculated the permanent dipole moment $p$ against the polarizability $\beta$. There is a clear linear relationship between $p$ and $\beta$, as shown in Fig. 10. This is coherent with recent

Fig. 9. $(z_e^0 - z_h^0)$ for S and P symmetries plotted as a function of applied electric field for $r_c = 8 \text{ nm}$.

Fig. 10. The permanent dipole moment $p$ versus the polarizability $\beta$.

Fig. 11. $p$ and $\beta$ plotted as a function of $h$ for $r_c = 8 \text{ nm}$. We show a linear relationship between them.
experimental observations done on individual quantum ring [25,26].

The linearity between $p$ and $\beta$ can be explained when we calculate the two quantities ($p$ and $\beta$) as a function of the QDs height $h$. A linear relationship between $p$ and $h$ and consequently between $\beta$ and $h$ is shown in Fig. 11. This result is in sound agreement with the experimental works showing that for a wide variety of QDs the linear relationship between the permanent dipole moment and the polarizability is rooted in the QDs-independent lateral extent of the electron and hole wave functions [25,26].

5. Conclusion

The electronic properties of the self-assembled InAs/GaAs QDs were investigated theoretically for the S, P and D symmetries. Calculations include the effect of a static electric field applied parallel to the cone axis. Our study of energy levels of the electron and hole and the electron–hole ground-state transition energies of the electron–hole are strongly supported by $z_0$ behaviour. The results found provide a plausible explanation of recent experimental observations carried out on InAs/GaAs QDs. Our study of the applied electric field and $r_c$ effects on the energy levels of the electron and hole enable us to have more information about correlation energies of excitonic complexes in these low-dimensional nanostructures. This will be our focus in a forthcoming paper.

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