InGaAs/GaAs quantum dot with material mixing

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ABSTRACT

An effective model for description of the electronic structure of the InGaAs/GaAs quantum dots (QDs) is presented. The model includes a single sub-band approach with an energy dependent electron effective mass and an effective potential which simulates the total effect of the strain and piezoelectricity. Based on our previous calculations for pure InAs/GaAs quantum objects which reproduce both the capacitance-gate-voltage (CV) experimental data and the ab initio calculations, we expand the model for the InGaAs quantum dots with significant Ga fraction. It is found that our model accurately describes the CV and photoluminescence (PL) from QDs with Ga fractions data up to 15%. We illustrate the accuracy of our model by comparing the calculated results and those obtained with the atomic pseudopotential model. We found that considerable difference of these models, appeared in the calculations, related to strength of the electron confinements.

Keywords: quantum dots, single carrier levels, optical properties

1 INTRODUCTION

The fabrication process of nano-sized self-assembled InAs/GaAs quantum dots (QD) and quantum rings (QR) may give the strained quantum structures with controlled geometrical properties [1,2]. There are experimental indicating [3,4,5] that a significant amount of Ga is incorporated into the quantum dots. This material mixing in the initially pure InAs quantum dots occurs during the growth process due to interdiffusions of the QD/substrate materials and can not be carefully controlled. The spatial distribution of the Ga fraction is not definitely known. Theoretical analysis [6-8], based on the effective mass kp-theory calculations, taking into account inter-band interactions, strain and piezoelectric effects by an ab initio manner, shows good ability to match experimental data. However, at present time the ab initio description is limited to the case of pure InAs QDs. In this work, we propose an effective model for strained InAs/GaAs quantum dots to study the character and magnitude of the changes to the energy spectrum of a single carrier arising from presence of the Ga fraction in QD. The model is based on a single sub-band approach for InAs/GaAs QDs with a energy dependent electron effective mass [9-11]. An additional potential V_s is included in the model to simulate the total effect of interband interactions, the strain and piezoelectricity. We have shown [12] that these effects may be taken into account in an effective manner through this approach. Using the model we have reproduced the results of the ab initio calculations [6-7] and the experimental capacitance-gate-voltage (CV) data [1]. We apply the effective model to study the InGaAs quantum dots with significant Ga fraction. Calculated results for QDs with 15% Ga fraction match both CV and photoluminescence (PL) data simultaneously. We illustrate the accuracy of our model by comparison of the calculated results with those obtained within the framework of the atomic pseudopotential model [13-16]. We found that the considerable difference of these models, displayed in the calculations, related to strength of the electron confinements.

2 EFFECTIVE MODEL

The described 3D heterostructure is modeled utilizing the kp-perturbation single subband approach with the energy dependent quasi-particle effective mass [8-11]. The energies and wave functions of a single carrier in a semiconductor structure are the solutions of the nonlinear Schrödinger equation:

$$\left(-(\nabla\frac{\hbar^2}{2m^*(r,E)}\nabla)+V(r)-E\right)\psi=0\,, \qquad (1)$$

where $V(r)$ is the band gap potential, proportional to the energy misalignment of the conduction (valence) band edges of InAs QD (index 1) and GaAs substrate (index 2). $V(r)=V_c$ inside the substrate, and $V(r)=0$ inside the quantum dot. The electron effective mass $m^*=m^*(x,y,z,E)$ is linearly dependent on energy for $0<E<V_c$ and varies within the limits of the QD/substrate bulk effective mass values. The magnitude of V_c is defined as $V_c=\kappa\left(E_{g,2}-E_{g,1}\right)$, where E_g is the band gap and the coefficient $\kappa<1$ is different for the conduction and valence bands. We use values for κ from Ref. [12]: κ^{CB}=0.54 and κ^{VB}=0.46. With experimental values $E_{g,1}$=0.42 eV, $E_{g,2}$=1.52 eV the band gap potential for the conduction band (valence band) is V_c=0.594 eV (V_c=0.506 eV). Bulk effective masses of InAs and GaAs are $m^*_{0,1}$=0.024 m_0 and $m^*_{0,2}$=0.067 m_0, respectively. Here m_0 is free electron

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mass. We use value of $m^* = 0.4\, m_0$ as the effective mass of heavy hole for both QD and the substrate. The presented band structure model is applied to "unstrained" InAs/GaAs structures. A realistic (*ab initio*) 3D model for QD has to take into account the band-gap deformation potential, the strain-induced potential, and the piezoelectric potential,

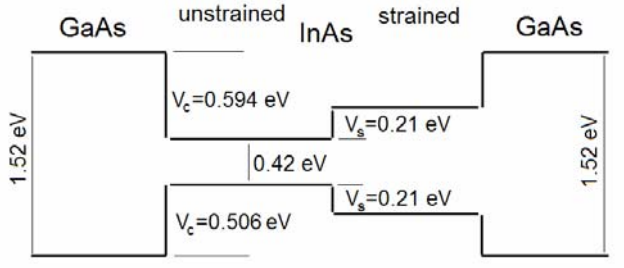

Figure 1. Band structure model for InAs/GaAs quantum dot.

in addition to the band-gap potential [6-8]. Alternatively one can take these potentials into account in an effective manner. In [17] we have introduced the potential V_s, which simulates the integrated effect of all QD potentials aforementioned. The effective potential V_s has an attractive character and acts inside the volume of the QD. The magnitude of the potential can be chosen to reproduce experimental data. This effective model was tested in [12] by comparison of the results of the calculations for QDs and quantum rings (QR) with the *ab initio* solution of the works [6-7] in which 8-band ***kp***-Hamiltonian has been used. It was shown that there is a good agreement of our results with those of the realistic calculations. The effective potential obtained from the comparison has value of 0.21 eV for the pure InAs/GaAs quantum dot. The band structure of our model is shown in Fig. 1. In the figure the effective model with potential V_s is denoted as "strained".

3 QD/SUBSTRATE MATERIAL MIXING

An InAs quantum dot having a semi-ellipsoidal shape embedded into the GaAs substrate is considered. Geometrical parameters of the QD are the height H, and circular base $b = 2R$. The previously reported geometry for experimental fabricated QD is defined by $b \cong 20$ nm, $H \cong 7$ nm [1, 18]. The cross section of QD considered in this section is shown in Figure 2.

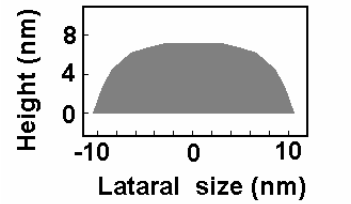

Figure 2. Cross section of semi-ellipsoidal shaped InAs quantum dot embedded in GaAs substrate

The effective potential V_s =0.31 eV was chosen for the interpretation of the capacitance-gate-voltage (CV) experiments of Ref. [1]. In the experiments the spectra of few electrons tunneling into QDs ware observed. There are two s and four p-shell electron levels below the GaAs conduction band edge [1, 18]. The first level of the d-shell is located above this threshold, but it can be observed by the Zeeman effect in a magnetic field.

	Ga, 10%	Ga, 15%	Ga, 20%	0.31 eV	Exp.
m^*_{QD}/m_0	0.055	0.056	0.058	0.057	0.057 $\pm$0.007 [1]
$\Delta E(e)$ $\Delta E(h)$	222 211	200 194	177 167	185 176	
$e_1 - e_0$ $e_2 - e_1$	47 55	46 54	45 51	46 52	44*[1]; 49*[18]
$h_0 - h_1$ $h_1 - h_2$	10 11	9 11	10 11	9 11	
E^c_{e0e0}	21.1	20.9	20.8	20.8	21.5[1]; 18.9[18]
E^c_{e0e1}	18.3	18.1	18.0	18.0	24[14]; 13.0[18]
E^c_{e1e1}	17.3	17.1	16.9	17.0	~18[14]
E^c_{h0h0}	25.2	25.0	24.8	24.9	
E^c_{e0h0}	22.8	22.7	22.5	22.6	
E^{ex}_{e0h0}	1060	1104	1154	1137	1098[14]
d_{00}	0.08	0.13	0.08	0.15	0.4 $\pm$0.1 [4]

Table 1. Calculated single electron (hole) energy-level spacing e (h), electron (hole) binding energy $\Delta E(e)$ ($\Delta E(h)$), electron-electron, electron-hole and hole-hole Coulomb energies $E^c_{\alpha\beta}$(α,β=e,h), excitonic band gap E^{ex} (in meV), exciton dipole moment d_{00} (in nm) and effective mass of the QD material for semi-ellipsoidal shaped InGaAs QDs (Ga fraction in %) embedded in GaAs.

With this potential the electron spectra calculation results in localization of the s-shell electron level with respect to the conduction band edge of the GaAs substrate, similar to that can be derived from the CV measurements (~180 meV). First order perturbation theory calculations [17] give a spectral picture that is in an agreement with the CV data. The effect of non-parabolicity, taken into account in our model by means of the energy dependent effective mass approximation, leads to a change in the electron effective mass of QD with respect to the bulk value. For the QD considered, the effective mass in InAs increases from the

initial bulk value of $0.024\,m_0$ to $0.057\,m_0$ (for p-wave electron), which is agrees with the experimental value of $0.057m_0 \pm 0.007m_0$, obtained in CV measurements by the Zeeman splitting of p-shell levels [1, 18]. From Table 1 one can see that the agreement between our results (the column "0.31eV") and the experiment data is satisfactory. The small numerical disagreement for few-electron spectrum obtained in [17] we have explained by the uncertainty of the QD geometry. In Ref. [12] we found that the small variations of the QD cross section can lead to quite significant changes of the electron level structure. In that work we did not consider the uncertainty which is related with material mixing of the quantum dot and substrate. From the experiments [3-5] it is evident that a significant quantity of Ga diffuses into the initially pure InAs quantum dots during the fabrication process. A more realistic model must take into account the material mixing. We assume that the effective model describes pure InAs/GaAs QD with the magnitude of the effective potential V_s =0.21 eV. This assumption is justified from comparison our model results with *ab initio* calculations [6-7]. Differences between the effective potential strengths, obtained for the experimental data (V_s =0.31 eV) and the *ab initio* calculation (V_s =0.21 eV), may be related to existence of the Ga fraction in the experimentally fabricated quantum dot. For V_s =0.21 eV potential we have performed calculations varying the Ga fraction in QD. Note that the effective electron mass and the band gap for $In_x Ga_{1-x} As$ material changes linearly with respect to the value of Ga fraction, assuming a homogenous distribution of Ga in the QD volume. The results of the calculations are listed in Table 1. The most reliable results we have gotten for the case of the 15% Ga fraction in QD. For this case we have also matched the experimental value of the transition energy for recombination of exciton pair (E^{ex}_{e0h0}).

4 COMPARISON WITH ATOMISTIC PSEUDOPOTENTIAL MODEL

The theoretical analysis of the electron state in the quantum dot with different Ga fraction distributions was also performed by A. Zunger and coworkers in Refs. [13-16] applying the atomistic pseudopotential approach. We have compared predictions of our effective model with the realistic calculations [6-7, 13] in Ref. [12] for the case of the pure InAs/GaAs quantum dots. As a result we concluded that the pseudopotential approach has strong electron confinement which does not allow us to reproduce the capacitance-gate-voltage measurement results simultaneously for QDs and QRs. In the present work, we continue the comparison of our approach and the atomistic pseudopotential treatment for the quantum dots having the material mixing with a substrate. We consider lens shaped InGaAs quantum dot embedded into GaAs substrate. The circular base of QD has a radius of 25.2 nm and a height varying from 2 nm to 10 nm. Calculations for a single electron and heavy hole energy levels are listed in Table 2 along with corresponding results of [14]. The Ga fraction in QD was chosen to be 15%. The height of QD was equal 7 nm and 3.5 nm for our calculations and the atomistic calculations, respectively. In spite of the quite different heights, one can see good agreement between the two calculations. The results of calculations are also in an agreement with experimental data given in Table 2. Coincidence of the calculations can be explained by the strong electron confinement of the atomic model. The decreasing of the vertical size of the quantum object can be compensated by the stronger confinement.

	[14]	Effective Model	Exp.
m^*_{QD}/m_0	--	0.055	0.057 ± 0.007 [1]
$\Delta E(e)$ $\Delta E(h)$	204 201	209 196	
$e_1 - e_0$ $e_2 - e_1$	52 60	42 43	44*[1]; 49*[18]
$h_0 - h_1$ $h_1 - h_2$	11 9	8 10	
E^c_{e0e0}	28	19.7	21.5[1]; 18.9[18]
E^c_{e0e1}	24	16.7	24[14]; 13.0[18]
E^c_{e1e1}	26	15.7	~18[14]
E^c_{h0h0}	30	23.8	
E^c_{e0h0}	29	21.2	
E^{ex}_{e0h0}	1083	1093	1098[14]
d_{00}	0.05	0.14	0.4 ± 0.1 [4]

*the quantization energy.

Table 2. Calculated result for lens-shaped InGaAs quantum dots with the Ga fraction of 15%. The notations identical to the those in Table 1. The QD has H =3.5 nm in [14] and H =7 nm in our calculations.

Note that the calculated exciton dipole moment d_{00} does not reproduce the experimental value as reported in Ref. [4]. This may indicate the existence of a non-homogenous distribution of the Ga fraction in vertical direction [4]. The agreement between other physical observables calculated by our model and the experimental data may be explained by the effective nature of our description. We also compared the effective model results along with the results obtained within the atomic pseudo-potential approach [14-16]. In these calculations the dependence of the transition energy of electron – heavy hole recombination for the InGaAs/GaAs quantum dots on the Ga fraction and vertical

size were obtained. The transition energy as a function of the Ga fraction value shown in Fig. 3 was calculated for two case of the QD height H =3.5 nm (open marks) and H =7 nm (solid marks). An interpretation of the results can be also based upon the stronger electron confinement in pseudopotential model compared with our model as well as in the *ab initio* model [12]. The compensation of such strong confinement can be obtained by increasing the Ga mixture in QD and/or decreasing the vertical size of QD. Note that the available experimental data for LP spectra rifer to the energy region from 1.0 eV to 14 eV. (see a review in [15]).

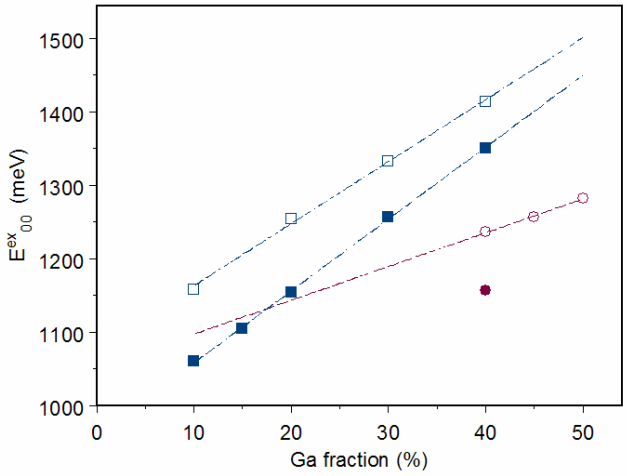

Figure 3. Transition energy of electron – heavy hole recombination (between the ground states) for various Ga fractions and the height H of the InGaAs/GaAs QD. The circles correspond to results of [15,16]. Our results are shown by the squares. The case H =3.5 nm (H =7 nm) correspond open (solid) marks. The straight lines are drawn only to lead the eye.

For the effective model calculations with $b \cong 20$ nm, this range approximately corresponds to quantity of the Ga fraction from 15% to 40%.

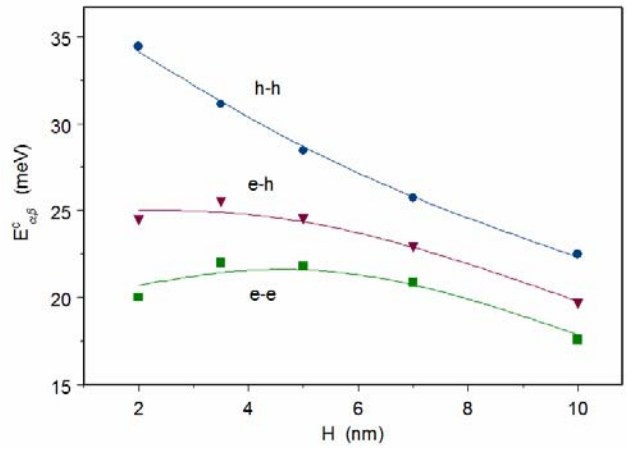

Figure 4. The height dependence of electron-electron, electron-hole and hole-hole Coulomb energies $E^c_{\alpha\beta}$ (α, β =e,h). The QD has a Ga fraction of 20%.

Continuing the comparison the effective model with molecular pseudopotential model, we present in Fig. 4 the result of our calculation of the Coulomb matrix elements for electron-electron, electron-hole and hole-hole interaction. In our calculations the lines corresponding to results for the matrix elements of different interaction do not cross, in contrast to results of the atomic pseudopotential model [16] for QD height of ~5nm. Note that the value of ~20 meV for the Coulomb energy of two electrons tunneling into s-shell of a QD was obtained from the CV experiments reported in [1,18]. Our calculations reproduce this value well, as one can see in Fig. 4.

5 CONCLUSION

In the framework of a effective model we have obtained satisfactory description of the CV and PL experimental data [1] for InGaAs/GaAs quantum dot with the Ga fraction about 15%. From the experimental PL data we found that available concentration of Ga in QD (with $b \cong 20$ nm) cannot be more than 40%. Analyzing the results calculated within the effective model and the atomic pseudopotential model we found considerable difference of both models related to strength of the electron confinements.

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