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2006 J. Phys.: Condens. Matter 18 1877

(http://iopscience.iop.org/0953-8984/18/6/005)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **18** (2006) 1877–1884 doi:10.1088/0953-8984/18/6/005

Effects of applied magnetic fields and hydrostatic pressure on the optical transitions in self-assembled InAs/GaAs quantum dots

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Received 25 October 2005
Published 24 January 2006
Online at stacks.iop.org/JPhysCM/18/1877

Abstract

A theoretical study of the photoluminescence peak energies in InAs self-assembled quantum dots embedded in a GaAs matrix in the presence of magnetic fields applied perpendicular to the sample plane is performed. The effective mass approximation and a parabolic potential cylinder-shaped model for the InAs quantum dots are used to describe the effects of magnetic field and hydrostatic pressure on the correlated electron–hole transition energies. Theoretical results are found in quite good agreement with available experimental measurements for InAs/GaAs self-assembled quantum dots.

Low dimensionality semiconductor heterostructures constitute attractive systems due to both their fundamental properties and their potential applications in optoelectronics devices [1–5]. Whereas quantum-well (QW) structures are already widely used in such devices, quantum-well wires (QWWs) and quantum dots (QDs) appear to be much more difficult to fabricate for these purposes. The formation of self-assembled quantum dots (SAQDs) by the Stranski–Krastanov growth mode in different material systems such as (Ga)InAs/(Al)GaAs or InP/GaInP heterostructures has been demonstrated successfully. Stopping the growth process in the initial state of formation of the nanostructure results in QDs free of defects and dislocations. Adler *et al* [6] have observed optical transitions from higher energetic dot levels in the photoluminescence (PL) spectra at different excitation levels in InAs/GaAs SAQDs. Their observed PL lines are in good agreement with calculated transitions between eigenstates obtained by solving the effective mass Schrödinger equation in cylindrical coordinates. Additionally, in their very simple model they have found only one quantized electronic level in the QD. Strain modifies the band structure via the deformation potentials, and the shift of the conduction band is proportional to the hydrostatic strain [7]. By using the solution of the 6×6 strain Hamiltonian for the valence band, Grundmann *et al* [8] have clarified the nature of the experimental absorption and luminescence spectra of self-organized InAs/GaAs QDs. Through a model calculation, they have also demonstrated that for those structures whose dimensions are of the order of 100 Å, only one quantized electronic level exists in the dots. A study of the temperature dependence of the PL emission from InAs QDs in a strained $Ga_{0.85}In_{0.15}As$ QW has shown [9] that the energy shift with the temperature generally follows the InAs band gap variation for temperatures up to about 200 K, and that when the temperature is further raised the QD peak redshifts faster than the InAs gap variation. The understanding of the magnetic field and pressure dependence of QD emission may be very important for building efficient lasers. Itskevich *et al* [10–12] have investigated the PL spectrum of self-assembled InAs QDs embedded in a GaAs matrix in magnetic fields up to 23 T and under hydrostatic pressure up to 70 kbar. They have found that the pressure coefficient for the dot emission line is 9.1 ± 0.2 meV/kbar, and have attributed the dependence on the pressure to such effects as the change in the InAs energy gap, a huge internal strain of the InAs dot, changes of the quantization energies of electrons and holes, and finally, on a minor scale, to changes in the Coulomb interaction energy and the small decrease of the dot size under compression. They conclude that a proper theoretical analysis of the electronic states of QDs is required to explain their pressure data. Also, quite recently, Ma *et al* [13] have made a thorough study of the PL spectra of InAs/GaAs SAQDs at 15 K under hydrostatic pressure up to 90 kbar. In this work we are concerned with a theoretical study of the effects of an external magnetic field and hydrostatic pressure on the PL spectra in InAs/GaAs SAQDs. Of course, a theoretical approach aiming at a proper understanding of the experimental findings [10–13] in InAs/GaAs SAQDs should include the size and shape of the SAQDs, stress effects, and valence-band mixing [14–16]. In the process of growth, stress effects are generated in the interfaces of the well and barrier and may be decomposed into hydrostatic and tangential components. The effect of both contributions to the conduction and valence bands at the Γ point leads to an additional splitting of the valence-band energies and, therefore, to different energy gaps between the conduction band and hh and lh bands [7].

From the experimental work by Itskevich *et al* [10, 11], the exact shape and dimensions of the InAs SAQDs are not known, and therefore we have chosen a simple treatment of the problem. The actual QDs have been modelled with an appropriate parabolic cylinder-shaped InAs/GaAs QD to describe the shape and stress effects in the dot and barrier layers, with the dimensions of the cylinder-shaped QDs given via a fitting of the theoretical e–hh transitions to the zero-stress experimental data from Itskevich *et al* [10, 12] and Ma *et al* [13]. We work in the effective mass approximation, and assume a cylinder-shaped InAs/GaAs QD model of radius R with an in-plane parabolic confinement potential. The radius of the dot is defined as $R = \sqrt{\hbar/\mu_{ex}\omega}$, where μ_{ex} is the hh–exciton reduced effective mass and $\hbar\omega$ is the lateral confinement energy. The effective mass excitonic Hamiltonian operator includes the kinetic energy of the conduction-band electron and the valence-band hole in the presence of the magnetic field, the screened electron–hole (e–h) Coulomb interaction and the QD confinement potential. The magnetic field is taken in the z direction, parallel to the growth axis. We write the exciton envelope wavefunctions $F_m(\vec{\rho}, z_e, z_h)$ as a linear combination of products [17–22] of single-particle $f_k(z_e)$ and $f_{k'}(z_h)$ solutions of the effective mass equation for electron or hole motion, respectively, along the z axis of a GaAs/InAs/GaAs QW of width h (which corresponds to the height of the model QD),

$$F_m(\vec{\rho}, z_e, z_h) = \sum_{k,k'} \psi^m_{k,k'}(\vec{\rho}, \varphi)\, f_k(z_e)\, f_{k'}(z_h), \tag{1}$$

where

$$\psi^{m}_{k,k'}(\vec{\rho}, \varphi) = \sum_{j} C^{m}_{k,k',j}\ \rho^{|m|}\ e^{im\varphi}\ e^{-\rho^2/\lambda_j^2}, \tag{2}$$

with the expansion in equation (2) made in a restricted set of Gaussian functions with appropriate λ_j length parameters [17–22]. Here we refer the reader to Barticevic *et al* [17–19] for details of the excitonic calculation approach, and note that essentially the same procedure was used in theoretical work [20, 21] on intraexcitonic transitions and magnetoabsorption spectra of GaAs–$Ga_{1-x}Al_xAs$ QWs, with results found in good agreement with experimental measurements. Moreover, the parabolic potential model QD adopted in the present study was used in a recent theoretical calculation [22] of excitons trapped in QDs/interface defects in narrow GaAs–$Ga_{1-x}Al_xAs$ QWs, with overall agreement with available experimental work. Also, one may note that a many-body approach used by Coli and Bajaj [23] in order to calculate the exciton binding energies in semiconductor QWs, over a large range of well widths, resulted in calculated binding energy values in very good agreement with those obtained by using a variational approach [24].

In what follows, the band offsets of the strained InAs/GaAs heterostructure were taken, for the conduction (valence) band, as 54% (46%) of the total band gap difference [6, 7]. The E_g hydrostatic, low temperature, pressure dependent band gap at the Γ point is [25]

$$E_g(P, T) = E_g^0 + \alpha P + bT^2/(T + c), \tag{3}$$

where E_g^0 is the $T = 0$, $P = 0$ energy gap, i.e., unstrained [26] $E_g^0(\mathrm{GaAs}) = 1.519$ eV, strained [6, 7] $E_g^0(\mathrm{InAs}) = 0.533$ eV, α is the pressure coefficient [27–29], $b(\mathrm{InAs}) = 2.76 \times 10^{-4}$ eV/K, and $c(\mathrm{InAs}) = 83$ K, whereas $b(\mathrm{GaAs}) = 5.405 \times 10^{-4}$ eV/K, and $c(\mathrm{GaAs}) = 204$ K. Other parameters we have used in the present calculations are taken for the stressed InAs QD and bulk GaAs, respectively, as follows [6, 14, 15]: conduction effective masses m_e = (0.040 and 0.0665), in-growth direction heavy-hole effective masses $m_{hh,z}$ = (0.59 and 0.377), in-plane heavy-hole effective masses $m_{hh,xy}$ = (0.035 and 0.112), and static dielectric constants [30] as ϵ = (14.6 and 12.35).

The hydrostatic pressure effects on the geometric dimensions of the InAs QD are obtained from the fractional change in volume [31]

$$\delta V/V = -3P(S_{11} + 2S_{12}), \tag{4}$$

where S_{11} and S_{12} are the compliance constants [31] given by

$$S_{11} = (C_{11} + C_{12})/[(C_{11} - C_{12})(C_{11} + 2C_{12})], \tag{5}$$

$$S_{12} = -C_{12}/[(C_{11} - C_{12})(C_{11} + 2C_{12})], \tag{6}$$

and C_{11} and C_{12} are the elastic constants [30, 32].

The magnetic field dependence of the excitonic spectrum is displayed in figure 1. We have achieved a good fitting with the experimental magnetic field dependent e–h transitions of Itskevich *et al* [10] by taking a cylinder-shaped QD of diameter and height of 94 and 10 Å, respectively. The size of the cylinder-shaped model QD is in fair agreement with the experimental work [10], as the authors comment that the sample was prepared by molecular beam epitaxy with a growth interrupt after deposition of 1.8 monolayers (MLs) of InAs that formed the QDs, and that they obtain a 60 Å in-plane spatial extension of the carrier wavefunction in the QD. We then used the above-mentioned cylinder-shaped InAs model QD to analyse the effects of hydrostatic pressure on the e–h transitions.

We first performed calculations for the pressure dependent uncorrelated e–h transitions, bound exciton and acceptor PL features in bulk GaAs (here we have used an

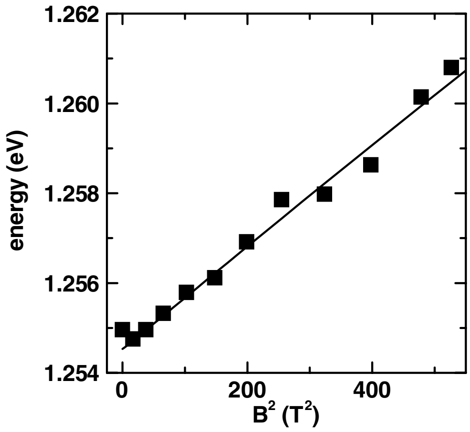

Figure 1. Energy position (squares) of the weighted PL line centre in InAs SAQDs embedded in a GaAs matrix, for a magnetic field applied perpendicular to the sample plane. The full line corresponds to the present theoretical results for correlated e–h transitions in parabolic potential cylinder-shaped (with diameter $d = 94$ Å and height $h = 10$ Å) InAs QDs, whereas experimental data (at $T = 10$ K) are from Itskevich *et al* [10].

unstrained [27] bulk α(GaAs) = 10.8 meV/kbar), with excellent agreement with experimental measurements [10], as seen in figure 2(a). We then studied, for parabolic cylinder-shaped InAs SAQDs, the effects of hydrostatic pressure on the correlated e–h transition energies (cf figure 2(b)), and the theoretical results (with unstrained [27] bulk α(GaAs) = 10.8 meV/kbar, and strained layer [13, 29] α(InAs) = 7.7 meV/kbar) are in quite good agreement with experimental measurements [11] up to ≈40 kbar. We notice that, at a hydrostatic pressure of the order of 9 kbar, the experiment [10] with InAs SAQDs shows an increase of the e–h transition energy of about 80 meV, whereas the theoretical increment [13, 29] in the InAs energy gap, in the case of a strained layer, is ≈70 meV. It is apparent, therefore, that the increase of the e–h transition energy with applied hydrostatic pressure is essentially associated with the variation of the InAs energy gap.

We next investigate the reliability of the present model calculation in the case of e–h excited states under hydrostatic pressure. Figure 3 displays further theoretical and experimental pressure dependent results of the e–h transition energies related to the ground and first few excited states in self-assembled InAs/GaAs QDs. Notice that the high pumping intensity PL experimental measurements [12] lead us to conclude that the observed lines correspond to e–h transitions from different excited conduction electron states in the InAs SAQD (nominal thickness 2.4 ML) sandwiched between two GaAs layers, with transmission electron microscopy indicating that the dots have a square base of length ≈150 Å and height ≈30 Å. Pressure dependent calculations of correlated e–h transitions with a parabolic potential cylinder-shaped (diameter $d = 106$ Å and height $h = 14$ Å) InAs QD are clearly in good agreement with the PL experimental data from Itskevich *et al* [12]. Also note that the theoretical geometric dimensions of the cylindrical InAs QD, found by fitting the dimensions to the first two zero-hydrostatic-pressure experimental measurements, are in fair agreement with the observed length and height of the QD if one considers that we are modelling a probably pyramidal dot by a simple cylindrical QD. Here, for the InGaAs strained layer grown on a GaAs substrate, we have followed the procedure by Frogley *et al* [29] and Ma *et al* [13], and used $\alpha = 7.7$ meV/kbar as the pressure coefficient.

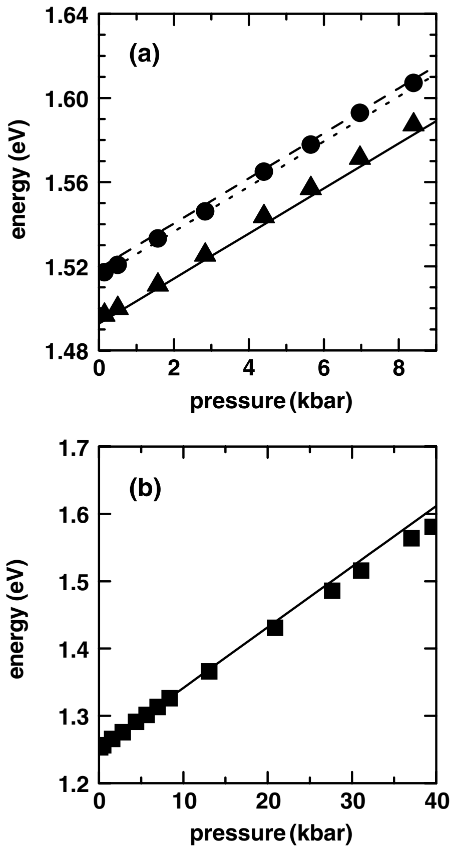

Figure 2. Theoretical pressure dependent (a) uncorrelated e–h (dashed line), bound exciton (dotted curve), and acceptor (solid line) PL features in bulk GaAs, and (b) correlated e–h transitions in a parabolic potential cylinder-shaped (with diameter $d = 94$ Å and height $h = 10$ Å) InAs QD (full curve). Full symbols are experimental data from Itskevich *et al* [10, 11] for bulk GaAs and InAs SAQDs, at temperatures $T = 4.2$ K for pressures up to 10 kbar [10] and $T = 12$ K otherwise [11].

In figure 4 we present results analogous to those in figure 3 but for the InAs/GaAs SAQDs studied by Ma *et al* [13]. Open symbols are the experimental data [13] obtained from PL measurements in a InAs/GaAs QD grown by molecular beam epitaxy in the Stranski–Krastanov mode with 2.5 ML InAs QD layers. In this particular case each InAs QD layer was covered by a 30 Å $In_{0.1}Ga_{0.9}As$ strain-reducing layer. Ma *et al* [13] have interpreted the observed PL features as follows: up-pointing triangles correspond to the direct gap of bulk GaAs at low temperatures, down-pointing triangles are the signal of the wetting layer, cross symbols in the low pressure regime (less than 40 kbar) are related to the e–h transitions of small-size InAs/GaAs QDs, and circles and squares correspond to PL features for the ground and first excited states of e–h transitions of large InAs/GaAs QDs. The theoretical pressure dependent direct gap—see equation (3)—of bulk GaAs is given by the dotted line in figure 4, and confirms the assignment by Ma *et al* [13]. Theoretical results for correlated e–h transitions for a parabolic potential cylinder-shaped (diameter $d = 114$ Å and height $h = 16$ Å) InAs/GaAs model QD are displayed as full lines in figure 4, and it is apparent that they describe quite well the observed ground and first e–h transitions assigned to large InAs/GaAs SAQDs. We note that we have

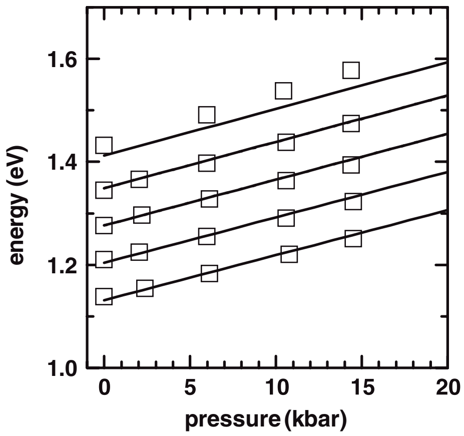

Figure 3. Theoretical (full lines) pressure dependent correlated e–h transitions in a parabolic potential cylinder-shaped (diameter $d = 106$ Å and height $h = 14$ Å) InAs QD. Open symbols are PL experimental data from Itskevich *et al* [12].

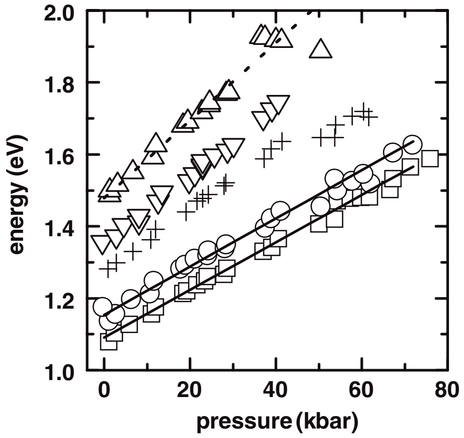

Figure 4. Theoretical (full lines) pressure dependent correlated e–h transitions in a parabolic potential cylinder-shaped (diameter $d = 114$ Å and height $h = 16$ Å) InAs QD. The dotted line is the theoretical pressure dependent direct gap (equation (3)) of bulk GaAs whereas symbols are experimental PL peak energies from Ma *et al* [13].

used the unstrained bulk $\alpha = 4.8$ meV/kbar for the InAs QD pressure coefficient [13, 28]. Additionally, we note that the pressure coefficients in figures 2 and 3 are in very good agreement with the PL features (cross symbols) Ma *et al* [13] assigned to small-size InAs/GaAs QDs.

Here we should mention that the exact shape and dimensions of the InAs/GaAs SAQDs in the PL experiments by Ma *et al* [13] are not known, as atomic force microscopy measurements were only made before the overgrowth of the InGaAs layer, and they argue that the QDs could undergo change in the dot size and shape during the overgrowth. From the theoretical point of view, we note that a careful study of the interpretation of the electronic structure of lens-shaped in contrast with pyramid-shaped InAs–GaAs SAQDs was performed by Williamson *et al* [33]. Focusing on the lens shape only, they examined the effect of changing the height and base of the assumed geometry, and argue that small changes in the geometry of the lens-shaped dot have only a small effect on the electronic properties that depend on the shape of the wavefunctions. Moreover, they mention that the effects of changing the geometry of the lens-shaped pure InAs dots on the single-particle energy levels may be qualitatively understood from single-band, effective mass arguments. We should add that theoretical work by Maksym and Chakraborty [34, 35], on the role of electron–electron interactions in quantum dots under a magnetic field, have concluded that, when the confinement potential is quadratic, the optical excitation energies of the many-body system are exactly the same as those of single-electron excitations. Also, the effect of the role of the Coulomb interaction between two electrons in a two-dimensional random potential was investigated by Talamantes and Pollak [36], whereas the interelectron interaction effects on a parabolic quantum dot subjected to an external magnetic field, taking into consideration the spin–orbit coupling, were recently studied by Chakraborty and Pietilainen [37, 38].

In conclusion, we have reported theoretical results on the applied magnetic field and hydrostatic pressure effects on the transition energies for e–h pairs in InAs/GaAs SAQDs. We stress that the exact shape and dimensions of the InAs SAQDs reported in the experimental work [10–13] are not known, and, in that sense, we have used a quite simple model calculation, in the effective mass approximation and using parabolic potential cylinder-shaped InAs QDs, which is shown to describe quite well the general aspects reported in experimental measurements by Itskevich *et al* [10–12] and Ma *et al* [13].

Acknowledgments

This research was partially supported by the Millennium Scientific Iniciative/Chile (Condensed Matter Physics, grant P02-054-F), Fondo Nacional de Ciencias/Chile (grants 1020839, 7010429 and 7020839), Universidad Técnica Federico Santa Maria (internal grant), Colombian COLCIENCIAS (grants 1106-05-11498 and 1115-05-11502) and CODI-Universidad de Antioquia Agencies, and Brazilian Agencies CNPq, FAPESP, Rede Nacional de Materiais Naoestruturados/CNPq, and Millenium Institute for Quantum Computing/MCT.

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