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Excitons in coupled quantum dots: hydrostatic pressure and electric field effects

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The variational procedure, in the effective-mass and parabolic-band approximations, is used in order to investigate the effects of hydrostatic pressure and in-growth direction applied electric field on the exciton states in vertically GaAs–$Ga_{1-x}Al_xAs$ coupled quantum dots. We have found that when the symmetrical lengths and radius of each QD are comparable with the Bohr radius of the GaAs material and for a finite value of the applied electric field, the binding energy always diminishes with the length of the central barrier because the two carriers in the exciton are localized in two well defined different regions of the system. However, for the zero electric field the binding energy decreases with the barrier width from the limit value corresponding to the exciton confined in one quantum dot of volume V up to reach a minimum and then increases to a value which corresponds to that of the exciton confined in an isolated quantum dot of volume $V/2$. Additionally, we have found that the applied electric field caninduce that the lowest structure in the photoluminescence-peak energy transitions be associated to spatially indirect excitons, situation which in the zero limit of the electric field and independent of the dimensions of the two coupled dots, always corresponds to spatially direct excitons. The main hydrostatic pressure effect reveals an increasing in the exciton binding energy, without modifying the direct or indirect exciton regime, and a well defined rigid blue-shift in the photoluminescence peak energy transitions in the presence of an applied electric field.

1 Introduction Zero-dimensional semiconductor quantum dots (QDs), being artificial atoms, have attracted considerable attention [1–7]. By coupling two or more QDs, the fabrication of artificial molecules opens a new way to investigate the quantum phenomena over a wide range of configuration. Electronic coupling processes have been investigated in coupled dot structures and the formation of bonding and anti-bonding states has been observed in symmetric dot-pairs [2]. A one step growth process is desired in order to avoid non-radiative recombination defects caused by nano-fabrication procedures. Stranski–Krastanow growth of self-assembled quantum dots (SADs) can be vertically selfaligned and electronically coupled. Fox et al. [1] have studied electron-sublevel-anticrossing effects in coupled quantum wells, finding that the crossings are shifted to higher values of the electric field due to the electron–hole interaction. Vertically coupled quantum dots (CQDs) have drawn a lot of interest recently due to the promising applications in spintronics where individual spins can be controlled to produce massive information storage devices [3, 4]. Excitons and correlated electrons have been studied in CQDs [5–7] and quantum wells [8–10]. They have found an nonparabolic Stark effect for the exciton associated with the electron–hole interaction and their results prove that by varying the width of the barriers between QD layers one can influence both the structural properties and the optical characteristics. The fractal dimension method [11] and the variational method [12], in the effective mass approximation, have been used to calculate the in-growth direction applied magnetic field effects on the ground state wave function of the exciton trapped in In(Al)As–Ga(Al)As CQDs and the built-in electric field

effects on the exciton in vertically GaN/$Al_xGa_{1-x}N$ CQD heterostructure. A theoretical study of the Stark effect for an exciton confined in a pair of vertically CQDs by Szafran et al. [13] predicted the mechanism and signatures of the trion recombination, which later on was experimentally confirmed by Krenner et al. [14].

In the present work, we develop a theoretical study of the hydrostatic pressure and in-growth direction applied electric field effects on the exciton binding energy and photoluminescence (PL) peak energy in GaAs–$Ga_{1-x}Al_xAs$ CQD. The effective-mass and parabolic-band approximations have been incorporated within a variational procedure. In Section 2 we present the theory of the problem. Our results are presented and discussed in Section 3, and our conclusions are given in Section 4.

2 Theoretical framework The Hamiltonian of an exciton confined in a CQD, under in-growth direction applied electric field (F) and hydrostatic pressure (P), using the in-plane relative coordinates $\rho = |\boldsymbol{\rho}_e - \boldsymbol{\rho}_h|$, can be written as [15]

$$H = -\sum_{i=e,h} \frac{\hbar^2}{2m_i^*}\left[\frac{\partial^2}{\partial\rho_i^2} + \frac{1}{\rho_i}\frac{\partial}{\partial\rho_i} + \frac{\rho^2 \pm (\rho_e^2 - \rho_h^2)}{\rho_i \rho}\frac{\partial^2}{\partial\rho_i\partial\rho}\right] + \sum_{i=e,h}\left[-\frac{\hbar^2}{2m_i^*}\frac{\partial^2}{\partial z_i^2} \pm eFz_i + V_i(\rho_i, z_i)\right] - \frac{\hbar^2}{2\mu}\left[\frac{\partial^2}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial}{\partial\rho}\right] - \frac{e^2}{\varepsilon\sqrt{\rho^2 + (z_e - z_h)^2}}, \quad (1)$$

where ($i = e, h$) and ± stand for electrons and holes, m_i^* is the effective mass, μ is the reduced-mass, e is the absolute value of the electron charge, ε is the GaAs dielectric constant, and the $V_i(\rho_i, z_i)$ is the electron/hole CQD confining potential. The hydrostatic pressure dependent dielectric constant and electron and hole effective masses are considered the same as in GaAs throughout the GaAs–$Ga_{1-x}Al_xAs$ CQD [16]. The electron and hole effective masses are given, respectively, by

$$m_e^* = \left[1 + \frac{15020}{E_g(P)} + \frac{7510}{E_g(P) + 341}\right]^{-1} m_0 \quad (2)$$

and

$$m_h^* = (0.45 - 0.1\times10^{-3}\ \text{kbar}^{-1}\ P)\, m_0\,, \quad (3)$$

where $E_g(P)$ is the bulk GaAs

$$E_g(P) = (1519 + 10.7\ \text{kbar}^{-1}\ P)\ \text{meV} \quad (4)$$

and m_0 is the free electron mass. The dielectric constant is obtained from

$$\varepsilon = 12.83\exp(-1.67\times10^{-3}\ \text{kbar}^{-1}\ P)\,. \quad (5)$$

In the model we consider two vertically coupled cylindrical QDs of radius R and lengths L_1 (QD1) and L_2 (QD2) separated by a central barrier of length L_b, i.e.,

$$V_i(\rho_i, z_i) = \begin{cases} 0 & \text{if} \quad \rho_i \le R, -L_1 - \frac{L_b}{2} \le z_i \le -\frac{L_b}{2}\,, \\ 0 & \text{if} \quad \rho_i \le R, +\frac{L_b}{2} \le z_i \le +\frac{L_b}{2} + L_2\,, \\ V_0(P) & \text{if} \quad \rho_i \le R, -\frac{L_b}{2} \le z_i \le +\frac{L_b}{2}\,, \\ \infty & \text{elsewhere}\,. \end{cases} \quad (6)$$

The potential $V_0(P)$ for electrons and holes is given by

$$V_0(P) = Q_C\left[1360x + 220x^2 + (0.8 - 1.3x)\ \text{kbar}^{-1}\ P\right]\text{meV}\,, \quad (7)$$

where x (= 0.3 in this work) is the alloy concentration and $Q_C = 0.6$ for electrons and 0.4 for holes. The dimensions of the structure (L_1, L_2, L_b, and R) are obtained by the fractional change in volume, which for the zinc-blende crystal of volume V is given by [17]

$$\frac{\delta V}{V} = -3P(S_{11} + 2S_{12})\,, \quad (8)$$

where S_{11} ($= 1.16\times10^{-3}$ kbar^{-1}) and S_{12} ($= -3.7\times10^{-4}$ kbar^{-1}) are the compliance constants of GaAs [16].

The exciton eigenfunctions may be obtained within a variational scheme with a trial wave function as

$$\Phi(\boldsymbol{\rho}_e, \boldsymbol{\rho}_h, z_e, z_h) = f(z_e)F(z_h)\,J_0\left(\frac{k_1\rho_e}{R}\right)J_0\left(\frac{k_1\rho_h}{R}\right)e^{-\lambda r}\,, \quad (9)$$

where λ is a variational parameter, J_0 (with $J_0(k_1) = 0$) is the usual spherical Bessel function of order zero, and $f(z_e)$ and $F(z_h)$ are, in general, linear combinations of the z-dependent part of the electron, $\sum_i a_i^{(e)} f_i(z_e)$, and hole, $\sum_j b_j^{(h)} F_j(z_h)$, eigenfunctions of the z_e and z_h dependent Hamiltonian neglecting the Coulomb interaction (see Ref. [18], and references therein). The coefficients $a_i^{(e)}$ and $b_j^{(h)}$ of these linear combinations are also variational parameters satisfying the usual normalization conditions [1]. The non-correlated $f_i(z_e)$ electron and $F_j(z_h)$ hole eigenfunctions may be obtained via the method by Xia and Fan [19] of expansion in terms of sine functions.

3 Results and discussion In Fig. 1 the results are presented for the exciton binding energy in a GaAs–$Ga_{0.7}Al_{0.3}As$ vertically CQD as a function of the length of the central barrier. In Fig. 1(a) because the sym-

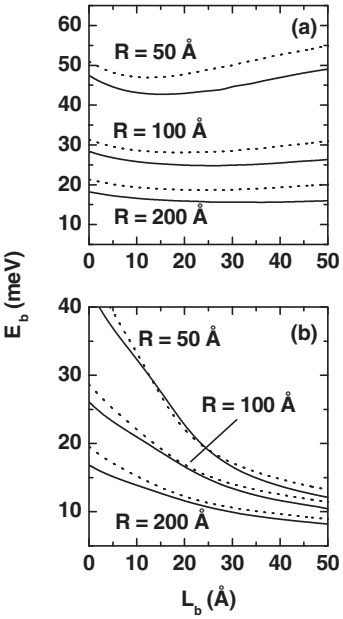

Figure 1 Exciton binding energy in a GaAs–$Ga_{0.7}Al_{0.3}As$ vertically coupled quantum dots as a function of the length of the central barrier. Results are for different radius of the structure with $L_1 = L_2 = 50$ Å. Solid lines are for $P = 0$ whereas dotted lines are for $P = 50$ kbar. In (a) $F = 0$ and in (b) $F = 100$ kV/cm.

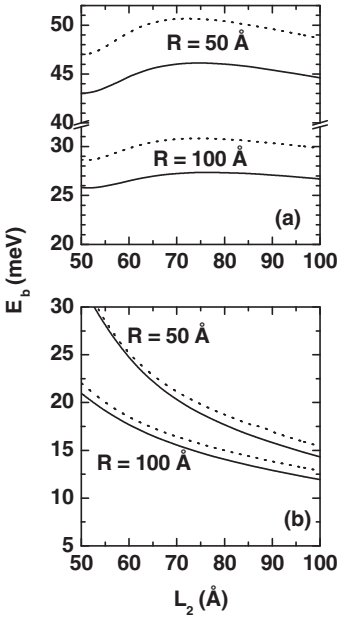

Figure 2 Exciton binding energy in a GaAs–$Ga_{0.7}Al_{0.3}As$ vertically coupled quantum dots as a function of the QD2 length. Results are for different radius of the structure with $L_1 = 50$ Å and $L_b = 10$ Å. Solid lines are for $P = 0$ whereas dotted lines are for $P = 50$ kbar. In (a) $F = 0$ and in (b) $F = 100$ kV/cm.

metry of the modeled system, the exciton is always in the direct spatial regime and the binding energy goes from the value corresponding to one isolated QD with length 100 Å, to one isolated QD with length 50 Å. In Fig. 1(b), due to the applied electric field, for $L_b = 0$ the electron and hole are in the same QD region but polarized, corresponding to one spatial direct exciton. For increasing L_b values the electron and hole are localized in separated dot regions. The system is not totally in the indirect exciton regime because the length of the each QD is too small with respect to the exciton Bohr radius which gives essentially the extension of the 1s-like wave function. Also, it can be seen that the hydrostatic pressure increases the exciton binding energy mainly due to the reduction with pressure of the dielectric constant.

In Fig. 2 results are presented for the exciton binding energy in a GaAs–$Ga_{0.7}Al_{0.3}As$ vertically CQD as a function of the length of the QD2. In the Fig. 2(a), for $L_2 = 50$ Å the system is symmetrical and it corresponds to a direct exciton in a QD of length 50 Å. When L_2 grows, it breaks the symmetry and simultaneously the wave functions of the electron and the hole begin to move toward the region of the QD of larger length. This movement is not sudden and again originates a direct exciton but with the wave functions of the two particles in the region of the same dot. The effects of the central barrier and of the dot of length 50 Å disappear for large values of L_2 (>200 Å). The appearance of the applied electric field, Fig. 2(b), polarizes the excitonic system, the hole moves toward the dot of height L_1 and the electron remains confined in the dot of height L_2 giving origin to an indirect exciton. Again, and for the same reason discussed in Fig. 1, the binding energy increases with the hydrostatic pressure.

In Fig. 3 we display the results for the PL-peak energy transition in GaAs–$Ga_{0.7}Al_{0.3}As$ vertically CQD as a function of the hydrostatic pressure. Results are for different values of the radius of the structure and applied electric field. The lengths of the two QD and of the central barrier are depicted. For $F = 0$, the wave functions for the first electron and hole confined states are mainly localized in the QD2 region and for it the showed PL-peak curves, in Fig. 3, all of them correspond to direct exciton transitions. Because the direction of the applied electric field, in the

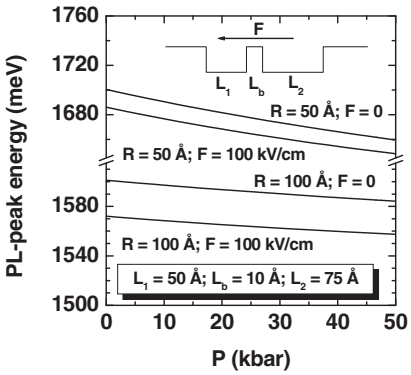

Figure 3 PL-peak energy transitions in a GaAs–$Ga_{0.7}Al_{0.3}As$ vertically coupled quantum dots as a function of the hydrostatic pressure. The pressure dependence of the GaAs band gap has been subtracted by using 10.7 meV/kbar as a pressure coefficient.

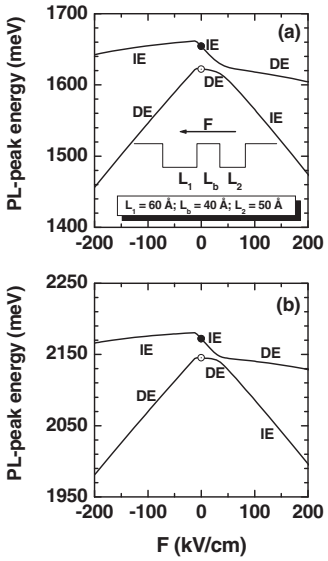

Figure 4 PL-peak energy transitions in a GaAs–$Ga_{0.7}Al_{0.3}As$ vertically coupled quantum dots ($R = 50$ Å) as a function of the applied electric field. In (a) the results are for $P = 0$ whereas in (b) $P = 50$ kbar. Open and full dots correspond to the PL-peak energy transitions at $F = 0$. The results are shown for direct (DE) and indirect (IE) exciton transitions.

case of $F = 100$ kV/cm, the electron wave function is pushed against the infinite barrier in $z = (L_b/2) + L_2$. Additionally, associated with $L_1 < L_2$ and due to the infinite barrier in $z = -(L_b/2) - L_1$, the hole wave function is mainly distributed in the region $+(L_b/2) \le z \le +(L_b/2) + (L_2/2)$. As a consequence, for $F = 100$ kV/cm the PL-peak energy transitions are also in the direct exciton regime. We note that there are not effects on the pressure coefficient of the PL-peak energy associated with the applied electric field. This is mainly due to the infinite external barriers in $z = -(L_b/2) - L_1$ and $z = +(L_b/2) + L_2$. Because of the finite barrier in $\rho = R$, we observe a decreasing in the pressure coefficient with the decreasing of the radius of the CQD structure. This effect has been previously reported by Venkateswaran et al. [20] in high-pressure studies of GaAs–$Ga_{0.7}Al_{0.3}As$ quantum wells.

In Fig. 4 we present our calculations for the PL-peak energy transitions in a GaAs–$Ga_{0.7}Al_{0.3}As$ vertically CQD as a function of the applied electric field. Two values of the hydrostatic pressure have been considered. Because the linear dependence with the pressure of the GaAs and the $Ga_{1-x}Al_xAs$ band-gap materials and the conduction effective mass, curves in Fig. 4(b) show essentially a rigid blueshift about of 524.8 meV with respect to the corresponding curves in Fig. 4(a), with small modifications (less than 3 meV) coming from changes in the exciton binding energy due to the applied hydrostatic pressure (see Fig. 1(a)). Because the asymmetry of the considered structure, at zero electric field the first confined electron and hole states correspond to the electron and the hole mainly localized in the QD1, namely direct exciton (DE), with the lowest PL-peak energy transition depicted by open dots in Fig. 4. The second confined electron state (orthogonal to the first one) corresponds to the electron localized mainly in the QD2 region. Because only one hole-confined state has been considered in our model, in this case the system corresponds to two particles localized in two well defined different regions of the system, namely indirect exciton (IE), with the highest PL-peak energy transition depicted by full dots in Fig. 4. With the increasing of the applied electric field (which direction is depicted in the inset) the effective gap of the heterostructure diminishes with the corresponding decreasing of all PL-peak energy transitions. The observed anticrossing, near to 50 kV/cm, corresponds to the resonance of the two first confined electron states in the CQD associated essentially with the first confined level, coming from both independent QDs. After the anticrossing the electric field pushes totally the electron wave function to the QD2 and the hole wave function remains in the QD1. This indirect exciton corresponds to the lowest PL-peak energy transition and the direct exciton transition corresponds to the higher energy values. For negative values of the applied electric field the electron and hole wave functions are pushed to the infinite barrier of the QD1 and to the central finite barrier which connect the two dots, respectively. Due to 1) the large dimensions of the central barrier, 2) the infinite potential at $z = (L_b/2) + L_2$, 3), the relative small z-dimension of the QD2, and 4) the fact that the hole is less sensitive to the applied electric field because its large effective mass, up to $F = -200$ kV/cm the first confined exciton state corresponds to both the electron and hole in the QD1 (lower PL-peak energy) and the second confined state to the electron in the QD2 and the hole in the QD1 (higher PL-peak energy), and as consequence non anticrossing effects have been observed in the range -200 kV/cm $\le F \le 0$.

4 Conclusions Summing up, it has been presented a theoretical study of the binding energy and PL-peak energy transitions related to direct and indirect exciton states in GaAs–$Ga_{0.7}Al_{0.3}As$ vertically CQDs under hydrostatic pressure and in-growth direction applied electric field. A variational procedure within the effective mass approximation have been used. We have found that when the symmetrical lengths and radius of the each QD are in the order of magnitude of the Bohr radius (~100 Å in the GaAs) and considering the presence of applied electric field the binding energy always decreases when the length of the central barrier is increased because the two carriers in the exciton are localized in two well defined different regions of the space. For zero electric field and symmetrical CQD structure the density of probability to find both carriers either in the QD1 or QD2 is 0.5 and when the width of the central barrier increases the binding energy augments from the corresponding value of an exciton in a single QD of length $L_1 + L_2 = 2L$ to the binding energy of the exciton in a single QD of length $L_1 = L_2 = L$. For non-symmetrical sizes of the length of the two QDs and for zero electric field,

always the first exciton state corresponds to both carriers localized mainly in the region of the larger QD and for it the lowest PL-peak energy of the system is essentially in the direct exciton regime. Depending on the direction and strength of the applied electric field and of the lengths of the two QDs the first exciton state can be tuned to the indirect exciton regime. This possibility of tuning from the direct to the indirect regime between the first and second exciton states, when an external applied electric field is considered, is the responsible for the presence of the anticrossings in the PL-peak energy transitions in the CQD heterostructures.

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