Optical response of confined excitons in GaInAsSb/GaSb Quantum Dots heterostructures

R Sánchez –Cano\textsuperscript{1}, L Tirado-Mejía\textsuperscript{2}, N Porras-Montenegro\textsuperscript{3}, G Fonthal\textsuperscript{2}, H Ariza-Calderón\textsuperscript{2}

\textsuperscript{1}Departamento de Física, Universidad Autónoma de Occidente, A.A. 2790, Cali, Colombia
\textsuperscript{2}Laboratorio de Optoelectrónica, Universidad del Quindío, A.A. 4603 Armenia, Colombia
\textsuperscript{3}Departamento de Física, Universidad del Valle, A.A. 25360, Cali, Colombia

E-mail: rsanchez40@gmail.com

Abstract. The narrow–gap Ga\textsubscript{1-x}In\textsubscript{x}As\textsubscript{y}Sb\textsubscript{1-y} compounds are suitable materials for heterostructure devices operating in the infrared wavelength range. In these compounds grown by liquid phase epitaxy over GaSb single crystals, for \(x\) and \(y\) values in the range of 0.10 to 0.14 for both variables, the photoluminescence optical response at 12K is blue-shifted by 20 meV related to the photoreflectance response. We believe this behavior is due to possible higher electronic confinement in some places of the heterostructure, possibly formed in the interface during the growth process. In order to explain this behavior, in this work we study the exciton recombination energy in spherical Quantum Dots (QDs) on Ga\textsubscript{1-x}In\textsubscript{x}As\textsubscript{y}Sb\textsubscript{1-y}/GaSb, using the variational procedure within the effective-mass approximation and considering an electron in a Type I band alignment formed by two semiconductors with similar parabolic conduction bands. Our results are in good agreement with recent experimental results.

1. Introduction

In ternary III-V compounds semiconductor alloys A\textsubscript{1-x}B\textsubscript{x}D or AC\textsubscript{x}D\textsubscript{1-x} type the band gap energy and the lattice parameter are generally both function of a single composition parameter, so they cannot be selected independently. In the quaternary A\textsubscript{1-x}B\textsubscript{x}C\textsubscript{y}D\textsubscript{1-y} semiconductor alloys type, on the other hand, the two composition parameters, \(x\) and \(y\), allowed the band gap and the lattice parameter to be selected independently within the constraints of a given substrate system. Quaternary alloys are appropriate materials for heterostructure devices because they provide a natural means of tuning the lattice parameter. In particular, in the quaternary alloy GaInAsSb it is possible to tune the magnitude of band gap so as to work in the infrared wavelength range [1-5]. The samples used in this work were grown by the liquid phase epitaxy (LPE) technique; details of growing conditions have been published elsewhere [6]. In the growth process the substrate and the liquid solution are put together at a temperature below the liquidus temperature. As temperature decreases, the solid phase from the liquid solution will appear. If substrate and liquid solution are in contact at a temperature higher than the equilibrium temperature the liquid solution will dilute the substrate due to its subsaturate condition. And as the temperature decreases again, the formation of the layer is induced. Is in this process where quantum dots could be formed in the interface.
The optical characterization of a GaSb sample and two epitaxial films of Ga$_{1-x}$In$_x$As$_y$Sb$_{1-y}$, lattice matched to GaSb, were carried out by photoreflectance (PR) and photoluminescence techniques (PL) [7]. PL measurements were carried out in different zones of the samples exhibiting the same energy position. This behavior was also observed for PR measures.

A lot of attention has been dedicated to the study of low-dimensional heterostructures such as GaAs/GaAlAs, InGaAsP/InP and CdS [8-11] but there is a small number of works on the novel quaternary heterostructures.

We present a calculation of the binding energy ($E_{1b}$) and the ground state energy levels of heavy-exciton ($E_X$) in spherical quantum dots (QDs) with a finite height potential for dot radii in the range of 5 – 20 nm and several values of Indium concentration $x$ on Ga$_{1-x}$In$_x$As$_y$Sb$_{1-y}$ in the range of 0.10 to 0.14. The number 1 in the sub index corresponds to a ground state. We use the variational procedure within the effective-mass approximation, and we considered that electrons and holes are both confined within the same layers of Ga$_{1-x}$In$_x$As$_y$Sb$_{1-y}$. Excitons energies are calculated and compared with experimental data. These comparisons show that higher electronic confinements, such as QDs, are formed during the growth process of the film by LPE technique, in some zones of the heterostructure.

2. Theory
In this analysis it is considered an exciton, formed by an electron - hole pair, in a quantum dot system consisting of GaSb/GaInAsSb/GaSb such that the electrons and holes are both confined within the same layers of GaInAsSb.

In the effective-mass model treatment the Hamiltonian we use to describe the exciton localized in a quantum dot is given by:

$$
H = -\frac{\hbar^2}{2m_e} \nabla_e^2 - \frac{\hbar^2}{2m_h} \nabla_h^2 - \frac{e^2}{\epsilon_b(x)|r_e - r_h|} + V_e(r_e) + V_h(r_h)
$$

where $\epsilon_b(x)$ is the static dielectric constant, $m_e$ ($m_h$) is the effective mass of electron (hole) in Ga$_{1-x}$In$_x$As$_y$Sb$_{1-y}$ spherical quantum dot, $r_e$ ($r_h$) is the electron (hole) position relative to the center of the spherical QD and, $V_e(r_e)$ and $V_h(r_h)$ are the barriers height of the confining potential for electrons and holes respectively. The confinement potentials for the electron and hole, in the Hamiltonian of the system are given by:

$$
V_e(r_e) = \begin{cases} 0 & r_e, r_h < R \\ \Delta E_C & r_e, r_h > R 
\end{cases} \quad V_h(r_h) = \begin{cases} 0 & r_e, r_h < R \\ \Delta E_V & r_e, r_h > R 
\end{cases}
$$

where $R$ is the radius of the spherical QD, and $\Delta E_C$ and $\Delta E_V$ are the conduction and valence band offsets respectively.

The conduction and valence band offsets, $\Delta E_C$ and $\Delta E_V$, in the equation (2) are given by:

$$
\Delta E_C = \chi(GaInAsSb) - \chi(GalnAsSb) \quad ; \quad \Delta E_V = \Delta E_g - \Delta E_C
$$

where $\chi(GaInAsSb)$ and $\chi(GalnAsSb)$ are the electron affinity of the semiconductors, and $\Delta E_g$ is the band gap change.

We followed a variational approach to solve the Hamiltonian expressed in equation (1). Taking into account the spherical confining geometry, $V_e(r_e)$, $V_h(r_h)$ confinement potentials, and Coulomb interaction between the electron and hole we take the product of 1s-hydrogenic wave function as the trial wave function for the exciton [8-11]. The ground – state wave function is given by:

$$
\Psi(r_e, r_h) = \begin{cases} A(R - \alpha r_e)(R - \alpha r_h)e^{-\alpha(r_e + r_h)} & r_e, r_h \leq R \\ A(1-\alpha)^2 R^4 e^{-(\alpha-\beta)(R-r_e-r_h)} & r_e, r_h \geq R 
\end{cases}
$$
where \( A \) is the normalization constant of the wave function, and \( \alpha, \beta \) are the variational parameters to be determined after minimization of the total energy \( (E) \). With the boundary conditions:

\[
\left. \frac{1}{\mu_{\text{in}}} \frac{\partial \psi_{\text{in}}}{\partial r} \right|_{r=R} = \left. \frac{1}{\mu_{\text{out}}} \frac{\partial \psi_{\text{out}}}{\partial r} \right|_{r=R}
\]

where the subscript \( t \) is depending the electron or hole case and \( \mu_{\text{in}}^*, \mu_{\text{out}}^* \) are the reduced effective mass of the exciton inside and outside the spherical QD; we can express \( \beta \) by means of \( \alpha \) as:

\[
\beta = \frac{\gamma(\alpha + \alpha(1-\alpha)R) + \alpha - 1}{(1-\alpha)R} ; \quad \gamma = \frac{\mu_{\text{out}}^*}{\mu_{\text{in}}^*}
\]

The exciton energy \( E_X \) is found by minimizing the total energy \( E \)

\[
\left\langle \psi_{(r_e,r_h)} \right| H(\alpha, x, R) \left| \psi_{(r_e,r_h)} \right\rangle = E \left\langle \psi_{(r_e,r_h)} \right| \psi_{(r_e,r_h)} \right\rangle
\]

The variational binding energies of the \( 1s \)-excitons states \( (E_{1b}) \) are obtained by subtracting from the lowest electron \( (E_{1e}) \) and hole subbands energies \( (E_{1h}) \), the ground-state exciton energy \( (E_X) \) expressed in equation (7). These subbands energies, \( E_{1e} \) and \( E_{1h} \), are determined solving the transcendental equations numerically for finite spherical quantum wells. From these results it can be found the critical radius value required for the existence of a bound state [12].

3. Results and Discussion

The exciton binding energy, \( E_{1b} \), presents a maximum when plotted versus the radius of the exciton. \( E_{1b} \) increases as \( R \) is reduced until it reaches a maximum, and then decreases rapidly, as presented in figure 1. The value of \( R \) at which \( E_{1b} \) reaches a maximum is smaller for larger \( x \). Furthermore, we found that \( \langle R \rangle \approx 15 \text{nm} \) corresponds to the average radius for which the exciton binding energy is a maximum in the complete range of Indium concentration \( (0.11 \leq x \leq 0.15) \).

**Figure 1.** Binding energy of heavy-exciton of the ground state \( E_{1b} \), in a spherical GaSb/Ga\(_{1-x}\)In\(_x\)As/\( \text{Sb}_1-y-GaSb \) quantum dot as a function of the radius for three different Indium concentration: \( x=0.11, x=0.14, x=0.15 \).

**Figure 2.** Heavy-exciton energy of ground state in a spherical GaSb-Ga\(_{1-x}\)In\(_x\)As/\( \text{Sb}_1-y-GaSb \) quantum dot as a function of the radius for three different Indium concentration: \( x=0.11, x=0.14, \) \( x=0.15 \).

In figure 2 it is observed that the exciton energy increases as the radius decreases and approximates to a limiting value when the radius approaches to the critical radius. It corresponds to the particle radius
for which the electron is not confined \( (R \approx 6\text{nm}) \), while the hole is still confined. We found that for \( \langle R \rangle \approx 15\text{nm} \) and \( x = 0.14 \) the exciton energy is \( E_X = 0.65\text{eV} \). This value for the exciton energy is in agreement with the experimental photoluminescence response showed in figure 3. In materials where these quantum structures are present, PL spectra exhibit transitions in which they are involved. The difference between the band to band and QDs emission energy will be the responsible for the anomalous behavior presented by PL emission energy compared to PR oscillation energy. In figure 3 we show results obtained by PR and PL techniques [3] for a GaInAsSb sample. The photoluminescence optical response at 12K is blue-shifted by 20 meV related to the photoreflectance response. This behavior was not observed in GaSb single crystals as shown in figure 4.

**Figure 3.** Photoreflectance (solid squares) and photoluminescence (solid circles) spectra for sample GaInAsSb M51 measured at 12K; the photoluminescence peak is shift 20 meV related to the photoreflectance response [3].

**Figure 4.** Photoreflectance (solid squares) and photoluminescence (solid circles) spectra for GaSb single crystal measured at 12K; the photoluminescence peak and photoreflectance response are both in 0.81eV [3].

### 4. Conclusions
We have determined the ground-state energy and binding energy of heavy-excitons confined in GaSb-Ga\(_{1-x}\)In\(_x\)As,Sb\(_{1-y}\)-GaSb spherical quantum dots using variational procedure within the effective-mass approximation as a function of the quantum dot radius for several values of the Indium concentration \( (x) \).

We compared our theoretical results with the available experimental data and found good agreement. These comparisons show that higher electronic confinements, such as QDs, are formed during the process of growth of the film by LPE technique in some places of the heterostructure. This indicates that the assumption of finite barrier height and the introduction of the reduced effective mass of the exciton inside and outside the spherical quantum dot would be essential to achieve a good agreement between theory and experiment.

**Acknowledgments**
This work was supported by Universidad del Quindío, Universidad Autónoma de Occidente in Cali-Colombia, and the Center of Excellence for Novel Materials CENM under contract 0043-2005 with COLCIENCIAS.
References

[1] Shim K, Rabittz H and Dutta P 2000 *J. Appl. Phys.* **88** 7157
[2] Andreev I A, Il’inskaya N D, Kunitsyna E V, Mikhaĭlova M P and Yakovlev Yu P 2003 *Semiconductors.* **37** 949
[3] Stoyanov N D, Mikhaĭlova M P, Andreĭchuk O V, K D Moiseev, Andreev I A, Afraïlov M A and Yakovlev Yu P 2001 *Semiconductors.* **35** 453
[4] Popov A A, Sherstnev V V, and Yakovlev Yu P 1998 *Tech. Phys. Lett.* **24** 73
[5] Ru G, Zheng Y and Li A J 1995 Appl. Phys. **77** 6721
[6] Tirado-Mejía L, Ramírez J G, Gómez M E and Ariza-Calderón H 2006 *Brazilian J. Phys.* **36** 1070
[7] Tirado-Mejía L and Fonthal G, 2007 Private communication Laboratorio de Optoelectrónica, Universidad del Quindío, A.A. 460 Armenia Colombia
[8] Marín J L, Riera R and Cruz S A 1998 *J. Phys.: Condens. Matter* **10** 1349
[9] Porras-Montenegro N and Pérez-Merchancano S T 1992 *Phys. Rev. B* **46** 9780
[10] Laheld U E H, Pedersen F B and Hemmer P C 1995 *Phys. Rev. B* **52** 2697
[11] Greene R L, Bajaj K K and Phelps D E 1984 *Phys. Rev. B* **29** 1807
[12] Davydov A S 1965 *Quantum Mechanics*, Pergamo Press, 130