Physica B 447 (2014) 7–11

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Numerical investigation of the postgrowth intermixing effects on the optical properties of InAs/GaAs quantum dots

Manel Souaf [a], Mourad Baira [a], Bouraoui Ilahi [a,b,*], Larbi Saxi [a], Hassen Maaref [a]

[a] Université de Monastir, Laboratoire de Micro-optoélectronique et Nanostructures, Faculté des Sciences, 5019 Monastir, Tunisia
[b] Department of Physics & Astronomy, College of Sciences, King Saud University, 11451 Riyadh, Saudi Arabia

ARTICLE INFO

Article history:
Received 27 January 2014
Received in revised form
19 March 2014
Accepted 14 April 2014
Available online 2 May 2014

Keywords:
Quantum dots
Modeling
Intermixing
Photoluminescence linewidth

ABSTRACT

We report on a simple theoretical model allowing to investigate the rapid thermal annealing induced quantum dots intermixing and consequent inhomogeneous broadening. In this model, where the 3D Schrodinger equation has been solved, by the orthonormal wave function expansion method, for strained InAs QD, we assume a lens-shaped QD with a uniform indium composition and a constant aspect ratio during the intermixing process. The size and aspect ratio for as-grown InAs QD, have been estimated by matching the calculated interband optical transition energies to the experimental photoluminescence emission peaks from ground and excited states. The simulated results were correlated with photoluminescence data at various annealing temperatures. Keeping constant the QD aspect ratio, a good agreement has been found between experimental and calculated emission energies for different indium atomic diffusion lengths. Small QDs are found to be more sensitive to the intermixing than larger QDs. This study allows also to calculate the full width at half maximum (FWHM) and compare it with the experimental value. The theoretical calculations suggest that the origin of the inhomogeneous broadening is mainly related to the variation of the QDs size.

1. Introduction

Self-assembled InAs quantum dots (QDs) have been a subject of extensive research due to their interesting systems for fundamental physics and for the development of electronic and optoelectronic devices [1,2]. Post-growth compositional intermixing further offers new fields of applications including photonic integrated circuits and broadband light emitters and detectors [3,4]. The intermixing of self-assembled InAs/GaAs QD formed by the Stranski–Krastanov (S–K) growth method has been widely investigated by several methods [1,5,7,8]. While the emission energy from intermixed QD has been successively tuned over a wide range [7,8], an inhomogeneous intermixing has been reported to occur at a given intermixing degree [10–12]. Several theoretical and experimental works have been carried out to investigate the intermixing effects on the QDs optical properties [6–21]. The lack of information concerning the real shape and indium composition of the QDs presents a crucial part of the calculation [13].

Many numerical approaches dealing with the effect of interdiffusion on the optical properties of QDs have been reported [14]. Gunawan et al. [21] and Djie et al. [15] have used different shapes of QDs including pyramidal, cubical and spherical QDs; they used Fick's law and momentum space methods to calculate the electronic structure. Petrov et al. [16] rather presented a theoretical and experimental study using Fick's law. Maia et al. [17] and Osman et al. [18] proposed a model for lens-shape, suggesting that the indium concentration varies linearly from the bottom (100%) to the top (0%) of the intermixed QD.

In this paper we investigate theoretically the In–Ga interdiffusion effect on the InAs QDs inhomogeneous broadening. We propose a simple model for a lens-shaped QDs [6,12,16] allowing to reproduce and explain the observed impact of the post-growth intermixing on the PL properties of InAs/GaAs QDs. The proposed model is however general and can be used to investigate other QDs systems.

2. Theoretical approach

The calculation was carried out for a lens shaped InAs QD embedded in a large cylinder of a GaAs barrier material. In accordance with many previous theoretical studies, this geometry is the most realistic model to describe the three-dimensional confinement [1,5,21]. By modeling the intermixing effects, it is possible to evaluate its influence on the QDs parameters such as

* Corresponding author at: Department of Physics & Astronomy, College of Sciences, King Saud University, 11451 Riyadh, Saudi Arabia.
E-mail address: bilahi@ksu.edu.sa (B. Ilahi).

http://dx.doi.org/10.1016/j.physb.2014.04.051
0921-4526/© 2014 Elsevier B.V. All rights reserved.

composition, size, and optical transition energies. Accordingly, we have used a simple theoretical model allowing to calculate the optical transition energies at different annealing temperatures and to estimate the In diffusion length (see Fig. 1).

To calculate the carrier's confinement energies and their wave functions in InAs QD, the three-dimensional Schrodinger equation has been numerically solved using the orthonormal wave function expansion method in cylindrical coordinates [22]. The model can be described as follow:

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

where m^* is the electron or hole effective mass. E and $\psi(r,\varphi,z)$ are the quantized energy levels and the corresponding wave function, respectively. We have assumed the wave function as a linear combination of the expansion basis functions.

$$\psi(r,\phi,z)=\sum_{n,m,l}A_{nml}\psi_{nml}(r,\phi,z) \quad (2)$$

With n, m, and l are the positive integers, the orthogonal periodic functions ψ_{mnl} and the coefficients A_{nml} must be determined. To solve the exact single-particle states we calculate the matrix element of the Hamiltonian operator between individual orthogonal periodic functions. The matrix elements are evaluated from the Hamiltonian equation H:

$$H_{nmln'm'l'}=\iiint_\Omega\left(-\psi^*_{n'm'l'}\left(\nabla\frac{\hbar^2}{2m^*}\nabla\right)\psi_{nml}+\psi^*_{n'm'l'}V\psi_{nml}\right)d\,\Omega \quad (3)$$

The impact of strain on the carrier confinement due to the lattice mismatch between InAs and GaAs materials is adopted for the calculation. Hydrostatic and uniaxial strains are induced, it can be written as following [17,23]:

$$\varepsilon_h=(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}) \quad (4)$$

and

$$\varepsilon_b=\varepsilon_{zz}-\frac{1}{2}(\varepsilon_{yy}+\varepsilon_{xx}) \quad (5)$$

Where ε_b and ε_h are the hydrostatics and uniaxiale strain respectively.

$$\varepsilon_{xx}=\varepsilon_{yy}=\frac{a_{\mathrm{GaAs}}-a_{\mathrm{InAs}}}{a_{\mathrm{InAs}}} \quad (6)$$

$$\varepsilon_{zz}=-2\frac{C_{12}}{C_{11}}\varepsilon_{xx} \quad (7)$$

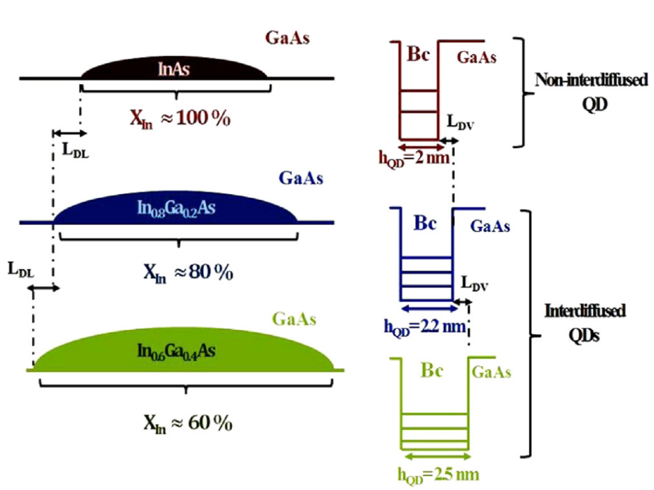

Fig. 1. Schematic model of a lens-shaped interdiffused InAs QD for different indium compositions.

the lattice mismatch between InAs and GaAs. The strained effect on conduction band is $\delta Ec=a_c{*}\varepsilon_h$ and for valence band is

$$\delta Ev=a_v(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})-\frac{1}{2}b(\varepsilon_b) \quad (8)$$

then, the strained band gap (Eg) can be written as following:

$$Eg_{\mathrm{InAs\_}str}=Eg_{\mathrm{InAs}-unstr}+\delta\,Ec-\delta\,Ev \quad (9)$$

Where $Eg_{InAs-unstr}$ is the unstrained InAs band gap energy.

However, during the annealing process, indium atoms out diffuse from the central core of InAs QD to the barrier due to the In–Ga interdiffusion effect. This may change considerably the chemical composition of QDs from pure InAs QD to a random $In_XGa_{(1-X)}As$ alloy. To simplify the calculation, we hypothesize a uniform In composition profile from the center to the border of the QD and we set a fixed aspect ratio during the intermixing process [17,18]. We define the lens shaped QD volume before annealing:

$$V_{QD}=\frac{h_{QD}\pi}{24}\left(4h^2_{QD}+3b^2_{QD}\right) \quad (10)$$

The corresponding QD volume after intermixing is as follows:

$$V'_{QD}=\frac{h'_{QD}\pi}{24}\left(4h'QD^2+3b'_{QD}2\right) \quad (11)$$

The indium composition can be derived as follows:

$$X_{In}=\frac{V_{QD}}{V'_{QD}} \quad (12)$$

where h_{QD}, h'_{QD}, b_{QD}, and b'_{QD} are the height and the base of non-interdiffused and interdiffused QDs respectively. This change of composition in the QD affects the lattice mismatch. As a result, the strain between GaAs and $In_XGa_{(1-X)}As$ QD is reduced witch influences the QD material's band gap, and carriers' effective masses. To take into account this change in our calculation, we have used Vegard's law:

$$a_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}}=xm_{\mathrm{InAs}}+(1-x)m_{\mathrm{GaAs}} \quad (13)$$

$$Eg_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}}=xEg_{\mathrm{InAs}}+(1-x)Eg_{\mathrm{GaAs}} \quad (14)$$

$$m^*_{In_xGa_{1-x}As}=x.m^*_{InAs}+(1-x).m^*_{GaAs} \quad (15)$$

3. Experiments

To correlate the proposed model with experiments we used single layer of self-organized InAs QDs sample grown by Molecular Beam Epitaxy (MBE) on semi-insulating (0 0 1) GaAs substrate at temperature of 500 °C. The QDs have been formed by depositing 2.4 ML of InAs followed by 50 nm GaAs barrier. For the intermixing procedure, the samples were protected by SiO_2 layer prior to the rapid thermal annealing for 30 s at the temperatures of 650 °C, 750 °C and 800 °C. More details on the growth process and characterization of these samples can be found elsewhere [7,28].

Fig. 2 presents the normalized 12 K PL spectra taken from the as-grown sample and samples annealed at different temperatures. The PL spectra have been recorded at 12 K for two excitation power densities showing that the observed multiple peaks arise from the ground and excited states emission energies. The PL peaks emission energies appear to be insensitive to the annealing process below 700 °C. With increasing the annealing temperature beyond 700 °C, the atomic interdiffusion considerably affects the QD PL properties [7,24,25]. The PL peaks position exhibits a blue shift around 180 MeV at T_a=800 °C. The energy spacing between the ground state and the first excited state emissions drops from 67 to 33 MeV [8]. The observed blueshift is accompanied by an