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INTERDIFFUSION OF IN/GA VERSUS ELECTRON COUPLING IN SELF-ORGANIZING DOUBLE QUANTUM DOTS OF INAS/GAAS GROWN BY MBE ON GAAS SUBSTRATES (001)

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ABSTRACT
The study addresses the quantum dot growth (QDs) of InAs/GaAs using the Molecular Beam Epitaxy (MBE) technique, focusing on the optical properties of self-organizing QDs, especially as a function of the thickness of the GaAs spacer layer and the number of layers of stacked QDs. The analysis of the samples reveals a bimodal behavior in the size distribution of the QDs, with emission of different energies associated with different sizes of the islands. The effect of layer stacking and the thickness of the spacer layer influence the optical emission shift, with blue-shift and red-shift effects observed, which are explained by the competition between electron coupling and the intermixing effect during growth. The use of thicker spacer layers leads to an increase in the homogeneity of the QDs and a shift to lower energies.

Keywords: Quantum Dots (QDs). Molecular Beam Epitaxy (EBM).

INTRODUCTION

In recent decades, a great development in the field of semiconductor physics has been achieved, mainly due to the rapid progress of semiconductor material growth techniques (PARKER, 1985; HERMAN et al., 1989), such as the Molecular Beam Epitaxy (EBM) technique. Among the various types of heterostructures that can be produced by the EBM technique are quantum dots (QDs). Semiconductor heterostructures are used in the manufacture of a wide variety of devices (MOZUME et al., 1999; YOON et al.., 1995; LI et al.., 1997, DINGES et. al.., 1994; CHELLES, 1995).

One type of heterostructure that has attracted great interest in recent years is the self-organizing quantum dot. The formation of QDs occurs, in this case, with the growth of a material with a different lattice parameter than the substrate lattice parameter; Thus, with the increase in the voltage of the network, the self-formation of three-dimensional islands that are the quantum dots occurs. This form of self-organized growth is known as Stranski-Krastanow (SK). Self-formation occurs spontaneously and depends on the conditions of growth, especially temperature. One of the challenges from an application point of view is to gain control of the

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sizes and distribution of the islands in the samples. Different sizes of QDs in a sample result in a bandwidth of optical emission, which limits the efficiency of the devices. The growth of samples with layers of vertically stacked QDs, separated by a spacer layer, known as stacked quantum dots, showed a vertical alignment of the QDs. The luminescence of stacked QDs is expected to have a narrower emission line width compared to single-layer samples due to the uniformity of QD size associated with the "stacking" effect. Stacked quantum dots have been studied for applications in infrared photodetectors and use in lasers (SILVA, 2008; LIU, 2003; JIANG et al.., 2004; LEDENTSOV, 2002; GRUNDMAN, 2000).

OBJECTIVE

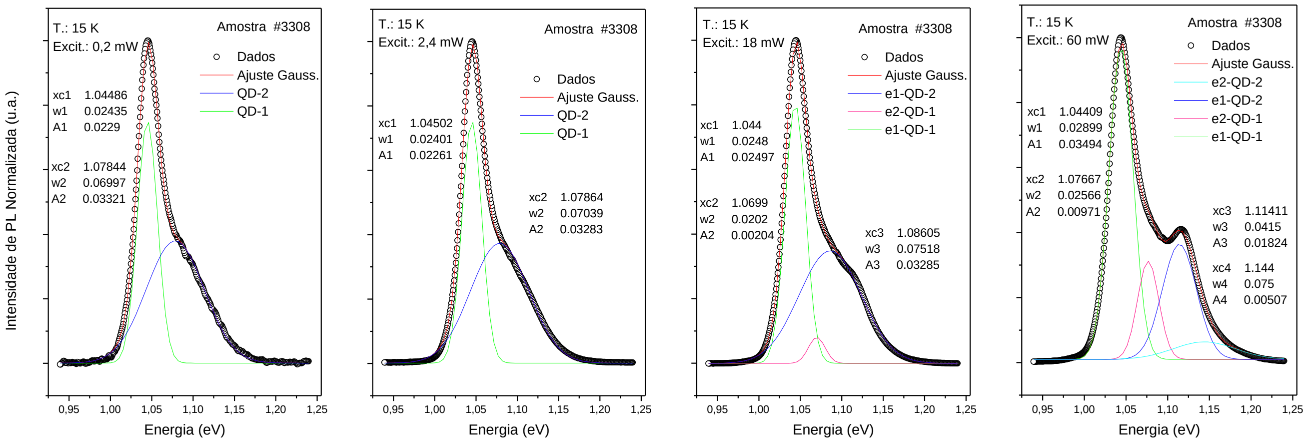
In this work, a set of samples of self-organized quantum dots of InAs/GaAs grown on GaAs(001) substrates was studied by the SK molecular beam epitaxy technique, with spacer layers of GaAs with different thicknesses, using the photoluminescence technique as a function of the intensity of the excitation laser and at low temperatures.

METHODOLOGY

The sample set studied is composed of six samples of QDs of InAs grown on GaAs (001). The reference sample 3308 has only one layer of InAs QDs, while in the 3309, 3310, 3311, and 3312 samples, two layers containing InAs QDs are grown separated by a GaAs spacer layer whose thickness d varies, respectively, as follows: 6 nm, 10 nm, 14 nm and 20 nm. Sample 3313 has a 14 nm spacer layer; however, in this sample, the structure of double layers of QDs was repeated 10 times, forming a kind of "super-network" of stacked QDs.

Figure 1 shows PL spectra obtained at low temperature for sample 3308, with excitation intensity ranging from 0.2 mW to 60 mW and with adjustments of the emission spectrum line form using Gaussian curves. The analysis of the spectra reveals a bimodal behavior for the size distribution of the QD islands in this sample. For low excitation intensities (0.2 and 2.4 mW), the experimental curves are well-fitted with two Gaussian curves, which are attributed to the transitions of the fundamental level of the QDs of the two families with different sizes (bimodal behavior).

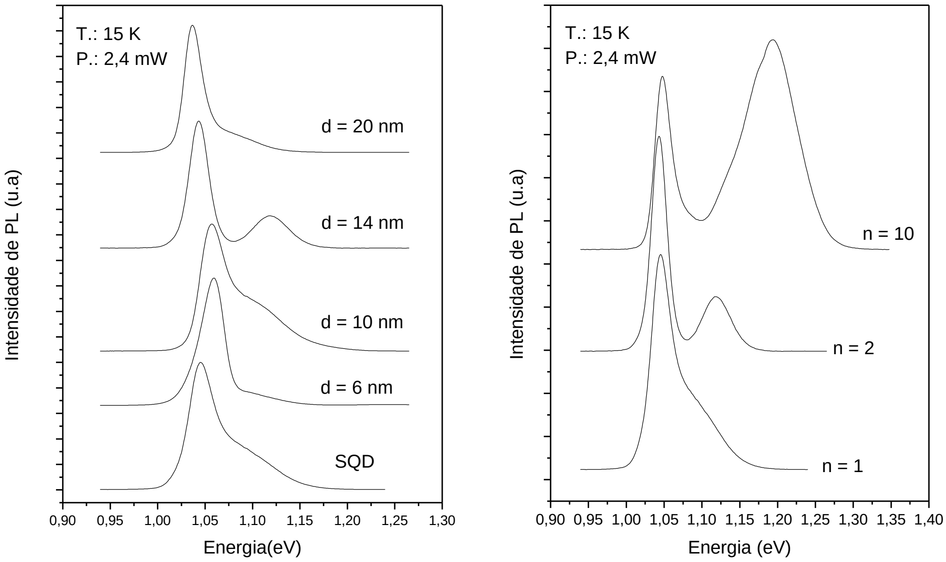
FIGURE A 1. PL spectra of the reference sample (3308) obtained at low temperature as a function of excitation intensity. The spectra are fitted by Gaussian curves.



The family with higher QDs emits in a region of lower energy (~1.05 eV), and the family with lower QDs emits in a region of higher energy (~1.08 eV). The bimodal behavior was also observed by Jung et al. (JUNG et. al., 2007) in PL analyses of stacked QDs of InAs/GaAs and QDs formed with other types of materials (ZUNDEL et. al., 1997). For the higher powers (18 and 60 mW), it is necessary to introduce one or two more Gaussians, depending on the power, to adjust the spectra appropriately. These additional curves are attributed to the transitions of excited levels associated with the two families of QDs. As the intensity of excitation increases, the lower energy states are populated, and recombinations involving the electron-hole pairs from the excited levels begin to occur, and this emission occurs at higher energies for both families of QDs.

Figure 2 shows the PL spectra of all samples, obtained at low temperature and as a function of the thickness of the spacer layer (Fig. 2(a)) and the number of layers (b). As the thickness of the spacer layer increases, from zero to 10 nm, there is a small displacement to the region of higher energy of the peak emission located around 1.05 eV. This transition is associated with the QDs of the first layer, and this small "*blueshift*" may be associated with a reduction in the average size of the QDs of the first layer from one sample to another. There is also a localized transition in a region of higher energy and that undergoes a shift to even higher energies ("*blueshift*") as *d* increases, even becoming well separated from the spectrum for *d* = 14 nm. We attribute this transition to the QDs of the second layer, although this transition does not occur with lower energy than the QDs of the first layer, as you would expect.

Figure 2: PL spectra obtained at low temperature (a) as a function of the thickness of the spacer layer and (b) the number of layers n.



According to the literature, the effect of stacking the layers leads to the formation of QDs with a larger size and with a more homogeneous distribution in the upper layer, which should cause the emission energy of these QDs to occur at a lower energy than the emission energy of the QDs of the lower layer. Therefore, the vertical electronic coupling between the QDs stacked in the different layers, together with the increase in the size and homogeneity of the QDs, should cause a red-shift of the emission to lower energies (SOLOMON et al., 1996). This "*red-shift*" depends, in principle, on the thickness of the spacer layer *d*. The greater the thickness, the smaller the electronic coupling and the smaller the "*red-shift*" effect and vice versa.

The results presented in Figure 2(a) for our samples are contradictory. Some studies have reported an "anomalous" displacement of emission to higher energies ("*blue-shift*") in stacked QDs, this displacement being attributed to the interfusion effect or "*intermixing*" between In/Ga of the QDs and the spacer layer, during the growth process (WANG et. al., 2007). The intermixing caused by the increase in the directed local voltage causes the emission energy to occur at a higher energy than expected for the quantum dots of the second layer, even though these QDs are larger than the QDs of the first layer. This effect has also been identified by other authors (LIPINSKI, 2000). The mixing of the GaAs barrier material in the InAs islands softens the containment profile and increases the energy levels in the QDs, thus increasing the transition energy (VELOSO, 2007). A similar effect was also identified in stacked QDs grown with other materials, such as Ge/Si (CHANG et al.., 2003).

The experimental results obtained in this work can be explained in terms of a competition between the effects of intermixing and electronic coupling. For spacer layers with thicknesses around 14 nm, the "*intermixing*" effect prevails, influencing the formation of the QDs of the upper layer, and a "*blue-shift*" is observed. This effect is reinforced in sample 3313, whose spacer layer is also 14 nm and has a "super-lattice" with 10 double layers of coupled QDs. For narrower spacer layers, this effect must be compensated for by the "*red-shift*" caused by the coupling of the QDs of the different layers. For sufficiently thick layers, both effects should be minimized, and an improvement in the homogeneity of the QDs, accompanied by a "*red-shift*", is observed for d = 20 nm.

FINAL CONSIDERATIONS

The reference sample, which contains only one layer of QDs, exhibits bimodal behavior. Even at low excitation intensities, two Gaussian curves are required to fit the PL spectra, which supports the assignment of bimodal behavior to this sample. The In/Ga intermixing effect shifts the emission of the QDs from the second layer to a region of higher energy due to the voltage formed by the In/Ga exchange during the QD growth process. This effect competes with the electron coupling effect, which, in turn, tends to shift the emission of the QDs from the second layer to the lower energy region of the spectrum. For the sample whose spacer layer thickness is 14 nm, the "intermixing" effect predominates. For the "super-lattice" sample of QDs with d =14 nm, an intensification of the peak located in the region of higher energy of the spectrum is identified, which is consistent with the attribution of the prevalence of the intermixing effect to this layer thickness. For samples whose spacer layers are narrower, this effect must be compensated by the "red-shift" caused by the coupling of the QDs of the different layers. For a sufficiently thick spacer layer (d = 20 nm), both effects should be minimized, and an improvement in the homogeneity of the QDs, accompanied by a "red-shift", is observed.

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