Influence of Annealing on the Optical and Electrical Properties of Multilayered InAs/GaAs Quantum Dots

A. J. Chiquito, Yu. A. Pusep, S. Mergulhão, Y. Galvão Gobato, and J. C. Galzerani

Departamento de Fisica, Universidade Federal de São Carlos, CP 676, 13565-905, São Carlos, SP, Brazil

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The characteristics of multi-layered InAs/GaAs self assembled quantum dots (SAQDs) annealed after the growth were here studied using a combination of capacitance-voltage (C-V) measurements, Raman scattering and photoluminescence (PL) spectroscopy. The combination of the results obtained with the three techniques, gave evidences that the annealing at 500^{o}C causes the sharpness of the SAQDs interfaces, while the annealing at 600^{o}C eliminated the SAQDs. However, the comparison with the case of single layered SAQDs, revealed a thermal stability of the last system even at an annealing temperature of 700^{o}C, thus confirming the role of the interlayer strain in the low temperature diffusion process.

The characteristics of multi-layered InAs/GaAs self assembled quantum dots (SAQDs) annealed after the growth were here studied using a combination of capacitance-voltage (C-V) measurements, Raman scattering and photoluminescence (PL) spectroscopy. For that, samples with different Si doping levels of the regions where the dots were grown were prepared. The combination of the results obtained with the three techniques, gave evidences that the annealing at temperatures close to the growth temperature causes the sharpness of the InAs/GaAs interfaces in the SAQDs system, while the annealing at 600^{o}C eliminated the SAQDs. However, the comparison with the case of single layered SAQDs, revealed a thermal stability of the last system even at an annealing temperature of 700^{o}C, thus confirming the role of the interlayer strain in the low temperature diffusion process.

The control of the homogeneity of the self-assembled quantum dots (SAQDs) is of fundamental importance when the application of these structures in technology is considered. Besides the attempts do attain the homogeneity through the deposition rate and the substrate temperature control, a post-growth annealing has also been attempted as a valid tool in recent works[1-4]. In this case, the homogenization was supposed to occur due to the atomic inter-diffusion process between the barriers and the islands (dots), which changes the sizes and the shapes of the dots. However, it is not yet clear how important are the relative modifications of the interfaces and the bulk of the SAQDs attained after the annealing.

In order to study the characteristics of multi-layered InAs/GaAs SAQDs structures annealed after the growth, we then used a combination of capacitance-voltage (C-V) measurements, and Raman and photoluminescence (PL) spectroscopies. The C-V measurements being sensitive to the electronic properties of the studied systems (as the PL spectroscopy does), in addition give information about their local electronic behavior, while the phonon Raman scattering is a widely recognized structural sensitive tool.

The samples were grown by molecular-beam epitaxy on (100) GaAs highly doped substrates. Five InAs layers with nominal thickness 2.3 monolayers (ML) separated by 15 ML of bulk GaAs were grown at 450oC. The transition from a streaked to a spotty high-energy electron diffraction (RHEED) pattern, observed after 1.8 ML thick InAs was deposited, indicated the formation of the 3D islands. After the growth of SAQDs, an undoped GaAs separating layer (25 nm) was grown, followed by an undoped GaAs/AlAs (1 nm/3 nm) superlattice, in order to increase the impedance of the samples. We used samples with two different Si doping levels of the GaAs regions where the dots were grown: sample A has $N_D^A = 1 \times 10^{17} cm^{-3}$ and sample B has $N_D^B = 1 \times 10^{17} cm^{-3}$. The post-growth annealing process was done in an argon atmosphere, for 20 min. at different temperatures.

For comparison, the effects of annealing were also studied in a single-layer SAQDs system.

The C-V measurements were made using a standard lock-in technique (with a SR530 Stanford dual-phase lock-in amplifier). Unpolarized back-scattering Raman spectra were performed with a Jobin Yvon

T64000 triple spectrometer supplied with a CCD detector cooled by liquid nitrogen, while for the PL measurements a Spex 500M single spectrometer with a photomultiplier tube was used. All the measurements were made at 10 K.

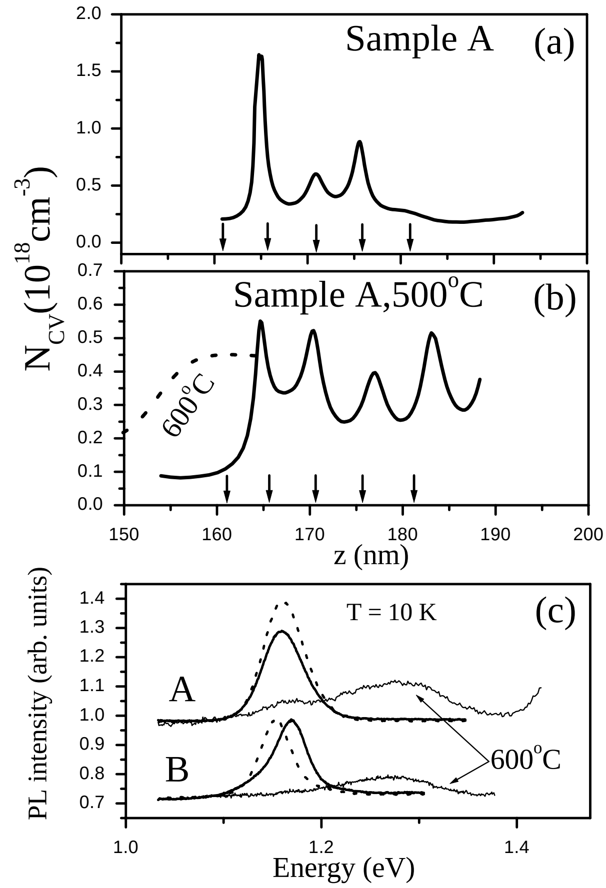

Figure 1. As-grown and annealed C-V profiles of the sample A are showed in panels (a) and (b). Panel (c) shows the PL spectra of the as-grown multilayer samples A and B (dotted lines) taken at 10 K. The corresponding spectra after the annealing at 500^{o}C are shown by full lines. The curves were shifted up for clearness and the excitation energy was 2.70 eV.

The spatial distribution of the electrons along the growth direction was calculated by the usual equations for the C-V profile determination [5]. Fig. 1(a) depicts the C-V profile of sample A, from which it is possible to notice the individual contributions of the InAs planes containing the dots, located at the following distances from the surface: 164 nm, 170 nm and 175 nm. The positions of the peaks are in accordance with the nominal localizations of the second, third and fourth InAs planes (indicated by arrows in Fig. 1). The first InAs layer does not contribute to the C-V profile (in this and also in sample B), because it was already depleted by the built-in field produced by the Schottky contact. After annealing at 500oC [Fig, 1(b)], the InAs planes containing the dots are better pronounced and more homogeneous. Similar results with the low temperature annealing were observed in sample B (not shown here). After the annealing at 600oC, the C-V profiles of both samples did not present any peculiarities related to the SAQDs, as depicted by the dotted line in Fig. 1(b) as an example; this is a first evidence that the dots have disappeared after the second annealing process (at 600^{o}C) in these multi-layered samples.

The corresponding PL spectra of the samples are depicted in Fig. 1(c). No significant influence of the low temperature annealing in the properties of the whole multilayer SAQDs system was observed, contrary to the significant modification in the individual properties of the layers measured by C-V. However, a strong modification of the PL spectra after annealing at a higher temperature (600^{o}C) was observed: the intensities of the lines associated with the SAQDs drastically decreased and a new blue shifted broad line appeared around 1.3 eV.

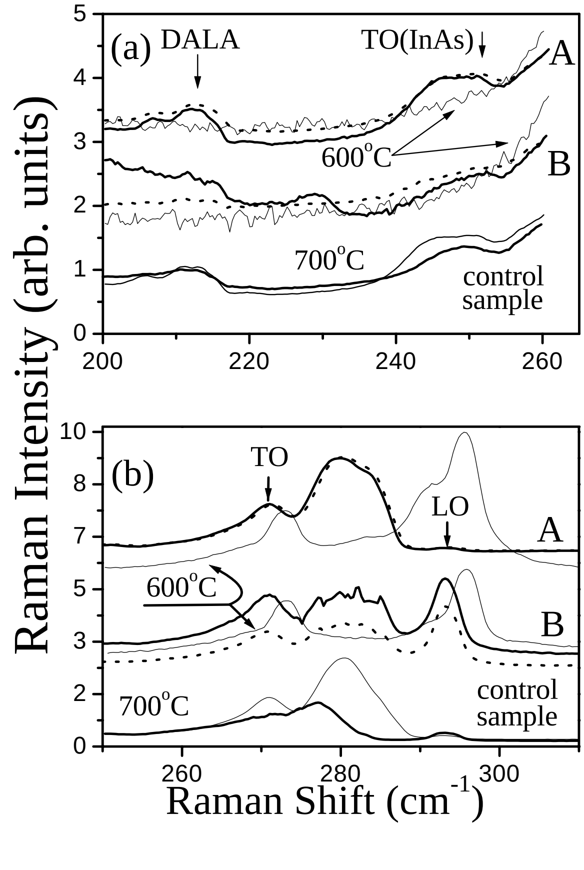

Figure 2. Raman spectra of the both multilayer and control samples (single layer SAQDs). Panels (a) and (b) show the Raman spectra measured in the frequency range of the InAs and GaAs phonons, respectively. The full and dotted lines represent the samples annealed at 500^{o}C and the as-grown sample, respectively; the thin lines were obtained after the annealing at 600^{o}C. In the bottom of the figure, the Raman spectra of both the as-grown and the annealed (700^{o}C) single layer SAQDs samples are presented.

The Raman spectra of the samples with the two different doping levels are shown in Fig. 2(a) for the frequency range of the InAs optical phonons. The full and the dotted lines represent the samples annealed at 500^{o}C and the as-grown sample, respectively; the thin lines were obtained after the annealing at 600^{o}C. In the botton of the figure, the Raman spectra of both the as-grown and the annealed (700^{o}C) single-layer SAQDs samples are presented. Following our previous experience, the line observed at 250 cm-1 was assigned to the InAs TO phonon, while no contribution was found at the frequencies where the InAs LO phonon was expected [6]. The weak line observed at 212 cm^{-1} is much probably a disorder activated longitudinal acoustic (DALA) phonon of GaAs. Since no significant shift of the bulk-line InAs phonon frequencies after annealing at 500^{o}C was observed, we conclude that the bulk structure of the SAQDs was not yet changed by the low temperature treatment.

We also performed the Raman spectra in the frequency range of the GaAs optical phonons [Fig. 2(b)]. The drawing lines have the same meaning as in Fig. 2(a). In this case, both the LO and the TO bulk phonons were clearly detected. In the as-grown samples, their frequencies are red-shifted due to the strain in the GaAs barriers. The absence of the bulk LO phonons of the GaAs substrate (despite the fact that the multi-layer SAQDs thicknesses are only 25 nm) is attributed to the inhomogeneity in these structures, that results in a strong elastic scattering of the light. As a support to this hypothesis, the LO phonon of the GaAs substrate appeared in the spectra after the high temperature annealing (600^{o}C), thus showing the formation of an alloy (without macro-defects like the quantum dots) that would prevent the elastic scattering of light to occur. In addition, a strong contribution of the GaAs-like optical phonons with large wave vectors, reflecting the one-phonon density of states, was detected between the TO and the LO lines, for the low temperature annealing. This defect induced Raman scattering drastically decreases after annealing at 600^{o}C; thus, it seems that the multi-layer SAQDs caused a strong violation of the Raman selection rules (that persists until after the low temperature annealing), while after the high temperature annealing mostly the zone-center phonons contributes to the scattering, as a further evidence that relatively homogeneous alloys were formed. As a matter of fact, after annealing at 700^{o}C, the SAQDs disappeared, and an $In_xGa_{1-x}As$ alloy was formed. As a result, the defect induced GaAs-like modes (DALA and the modes with large wave vectors) were eliminated, while the GaAs bulk-like phonons of the substrate now shows up. The shoulders detected at the low frequency sides of the LO Raman lines correspond to the GaAs-like phonons of a $In_{0,08}Ga_{0,92}As$ alloy, confirming the above related observation of the PL measurements: the shifted PL lines can be attributed to an $In_xGa_{1-x}As$ alloy with $x = 0,10$ formed as the result of the annealing.

So, it is worth noticing that the homogenization of the SAQDs and the increase of the localization of electrons, observed by the C-V measurements, was shown to imply in structural alterations, detected by the optical spectroscopies.

Contrary to the multi-layer case, the Raman spectra of the single-layer SAQDs revealed the presence of the InAs optical phonons and the GaAs-like defect induced modes associated with the dots even after the annealing at 700^{o}C, as evidenced in Figs. 2(a) and 2(b). This observation is confirmed by the PL measurements (spectra not shown here): the lines due to the dots are still observed after the high temperature annealing, although they are slightly blue-shifted and broadened. Thus, from the three techniques here employed, we could conclude that the low temperature heat treatment (at a temperature somewhat higher than the growth temperature) results in a drastic modification of the multi-layer SAQDs, while the single-layer structure revealed a thermal stability even at rather high temperatures. We believe that these observations confirm the role of the interlayer strain in the low temperature diffusion process.

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