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Raman study of interface modes subjected to strain in InAs/GaAs self-assembled quantum dots

Yu. A. Pusep and G. Zanelatto
Universidade Federal de São Carlos, 13565-905 São Carlos, SP, Brazil

S. W. da Silva
Instituto de Fisica, Universidade de Brasilia, 70910-900, Brasilia, DF, Brazil

J. C. Galzerani
Universidade Federal de São Carlos, 13565-905 São Carlos, SP, Brazil

P. P. Gonzalez-Borrero
Instituto de Fisica de São Carlos, Universidade de São Paulo, 13560-970 São Carlos, SP, Brazil

A. I. Toropov
Institute of Semiconductor Physics, 630090 Novosibirsk, Russia

P. Basmaji
Instituto de Fisica de São Carlos, Universidade de São Paulo, 13560-970 São Carlos, SP, Brazil

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Interface vibrational modes localized at the apexes of pyramidal InAs self-assembled quantum dots embedded in GaAs were observed by resonance Raman scattering. The comparison of the frequency positions of the interface modes with those obtained theoretically reveals a strong influence of the strain. The strain calculated for the InAs/GaAs dots satisfactorily explains the strain-induced frequency shifts obtained for the interface modes. It is important to notice that the interface modes observed in this study can be found in any corrugated interfaces containing tips and cusps where they can be localized. [S0163-1829(98)50428-6]

Self-assembled quantum dot structures have attracted much attention over the past years because of the possibility of obtaining defect-free systems confining electrons without processing by lithography and etching. As is well known, self-assembling of quantum dots (QD's) occurs during molecular beam epitaxy (MBE) growth of highly mismatched heterostructures.[1–3] Strain plays a crucial role in the process of QD formation providing a natural driving force which causes the transition from two-dimensional growth mode to the formation of pyramidal-shaped[4] or hemispherical[5] islands based on a thin wetting layer. This transition takes place above a critical coverage, for instance 1.7 monolayers (ML) for InAs on GaAs.[3] Therefore, the study of the strain can provide us with important information in order to understand and to control the growth of the self-assembled quantum dots.

A number of papers were devoted to the calculation of strain in such systems and to the analysis of the effects of strain on their electron and phonon spectra.[4,6–8] In the experimental field most attention was paid to the study of the electron structure (for instance, see Refs. 2, 9, and 10); however, knowledge about phonons is still poor,[4,11,12] although the study of phonons is recognized as a powerful tool to investigate the strain in crystals.

Furthermore, the self-assembled quantum dot system reveals a strongly corrugated interface and can serve as a good candidate to study the interface (IF) modes localized at the tips and cusps; the existence of such modes was predicted in Ref. 13.

Here we present an investigation of the optical phonons in the strained InAs/GaAs heterostructures containing self-assembled dots. The optical phonons of the GaAs barrier and of the InAs dots were detected by the resonance Raman scattering at the $E_0+\Delta_0$ gaps of GaAs and InAs.

The strained InAs/GaAs heterostructures were grown on the (001)-oriented GaAs substrates by molecular beam epitaxy via Stranski-Krastanov growth. Structures with and without 500 Å thick cap layers were grown under the same growth conditions; the first ones were studied by Raman scattering, while the second ones were characterized by an atomic force microscope (AFM).

The Raman measurements were performed at $T=8$ K using a Jobin-Yvon U-1000 double-grating spectrometer supplied with a conventional photon counting system. A jet-stream DCM dye laser and a Ti-sapphire laser both pumped with an Ar^+ ion laser was used for excitation. In order to avoid the photoluminescence background the measurements were realized in the cross-polarized $z(\underline{xy})\overline{z}$ configuration of Raman scattering with $x\|[110]$, $y\|[1\bar{1}0]$, $z\|[001]$. The topologies of the surfaces of the samples were examined *ex situ* by a Digital Instruments Nanoscope IIIa using the tapping mode.

The AFM images of two samples grown with different nominal thicknesses of InAs (2 and 3 ML's) and, as a consequence, with different densities of dots N_{QD} (2×10^{10} cm^{-2} and 5×10^{10} cm^{-2}, respectively) are depicted in Fig. 1 demonstrating the formation of the InAs dots. In the samples where the dots are well separated the typical base

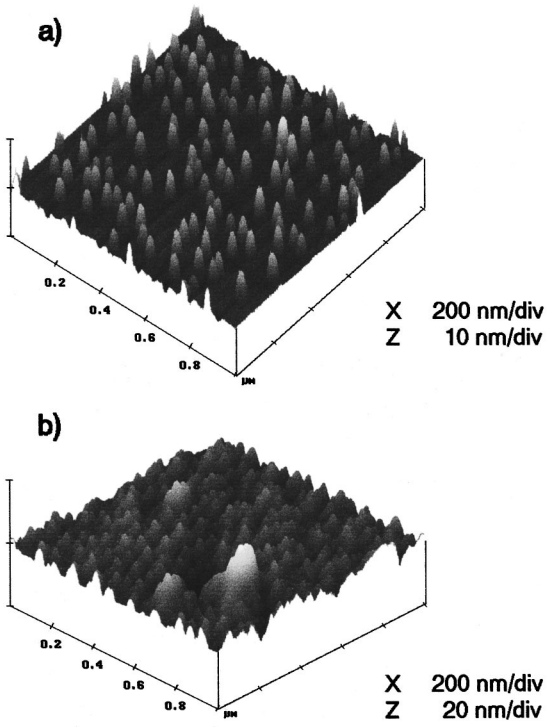

FIG. 1. The atomic-force microscope images of the InAs/GaAs heterostructures grown with nominal thicknesses of InAs: (a) 2 ML's ($N_{QD}=2\times10^{10}$ cm^{-2}), and (b) 3 ML's ($N_{QD}=5\times10^{10}$ cm^{-2}) presenting the formation of the InAs dots.

length is 36 nm with a height of 7 nm. The Raman spectra of samples with different densities of dots are shown in Figs. 2 and 3. Two lines, attributed to the TO and LO phonons in the InAs dots, were found at the excitation energy close to the

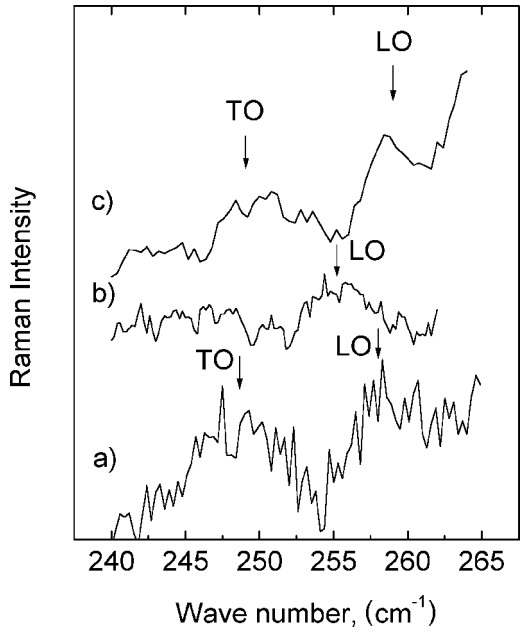

FIG. 2. The Raman spectra of the InAs/GaAs heterostructures grown with different nominal thicknesses of InAs: 2 ML's (a), 2.5 ML's (b), and 3 ML's (c), measured in the frequency range of the InAs optical phonons with the excitation energies 1.853 eV, 1.7 eV, and 1.853 eV correspondingly at $T=8$ K.

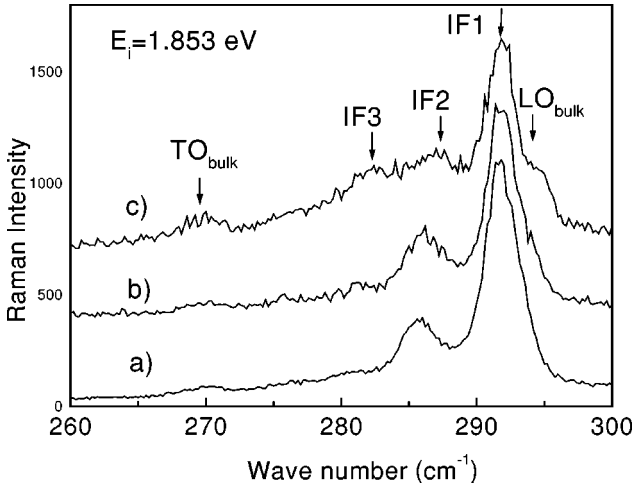

FIG. 3. The Raman spectra of the InAs/GaAs heterostructures grown with different nominal thicknesses of InAs: 2 ML's (a), 3 ML's (b), and 6 ML's (c), measured in the frequency range of the GaAs optical phonons at $T=8$ K with the excitation energy 1.853 eV.

$E_0+\Delta_0$ gap of the InAs quantum dots (1.7 eV as measured by photoluminescence in Refs. 14 and 15). Both the TO and LO phonon lines of InAs are strongly shifted due to the strain. The frequencies of the LO phonons found in the InAs dots around 255–258 cm^{-1} are in good agreement with the value measured by multiphonon relaxation (259.7 cm^{-1}) and with the calculated average frequency (258.9 cm^{-1}).[4] For the TO phonon we found a line at 250 cm^{-1}, while the calculations give the average frequency 242.4 cm^{-1}.[4] It is worth mentioning that the TO and LO frequencies of unstrained, bulk InAs are 218 and 243 cm^{-1}, respectively.[16]

In the frequency range of the GaAs optical phonons we observed five lines. The frequency positions of all these lines measured at different excitation energies are collected in Fig. 4. Two lines, at 295 and at 270 cm^{-1} which do not exhibit any dependence on the excitation energy are assigned to the LO and TO phonons of bulk GaAs, respectively; the inten-

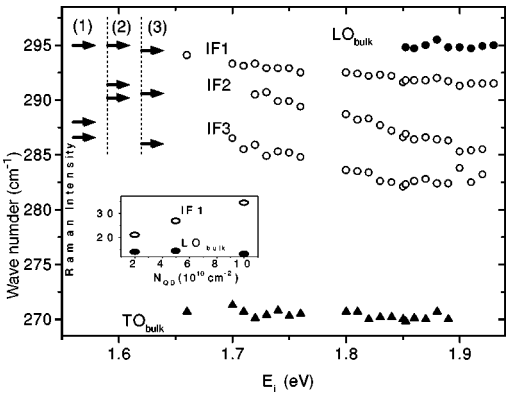

FIG. 4. The positions of the Raman lines in the spectra of the InAs/GaAs self-assembled quantum dots, whose spectra are shown in Fig. 3(c), measured at different excitation energies. The calculated frequencies of the interface modes are shown for a spherical dot (1), for an apex of a pyramid (2), and for an apex of a pyramid subjected to strain (3). The insertion shows the dependencies of the intensities of the lines on the densities of InAs dots (N_{QD}).

sities of these lines do not depend on the dot densities. The bulk LO phonon line reveals both the incoming and outgoing resonances at the energies (1.86 and 1.90 eV, respectively) found to be in good agreement with those measured in the bulk GaAs.[17] The intensities of the other three lines measured between the bulk LO and TO phonons of GaAs increase with the dot density; these lines reveal a dispersion with the excitation energy and they were attributed to the interface GaAs-like vibrational modes (IF1, IF2, and IF3 modes as they are labeled in Fig. 4). The behavior of the intensities of the first IF mode and of the bulk LO phonon is shown in the inset of Fig. 4; the IF1 mode was measured with the excitation energy $E_i = 1.845$ eV, while the bulk LO phonon was taken with $E_i = 2.4$ eV in order to avoid the masking by the IF modes.

The IF modes of quantum dots with different shapes were studied within the dielectric continuum approach in Ref. 13. It was shown that the dots with sharp tips (like the apex of a pyramid) support IF modes which are strongly enhanced in the vicinity of the tip. According to these calculations the IF modes localized at the infinitely sharp tips reveal a continuous vibrational spectrum limited by a maximum frequency, while those of the smoothed tip (as in real dots) are characterized by a discrete spectrum.

The frequencies of the first three GaAs-like IF modes calculated by the formulas and the data presented in Ref. 13 are shown in Fig. 4 by arrows. These calculations were fulfilled for a tip with the form of a ''spindle '' with an aspect ratio corresponding to a pyramid with equal base length and height; the sharpness parameter which characterizes the tip was taken as $R/a = 10^5$ (the significance of this parameter is explained in Ref. 13). For comparison, the frequencies of the IF modes calculated by the same formalism for a spherical dot are displayed in Fig. 4 as well. As it follows from the calculations, the first fundamental IF mode of the tip has the same eigenfrequency as the bulk LO phonon. According to Ref. 13, this mode can be found only in the barrier and vanishes within the dot. The high-index IF modes are grouped in a narrow frequency interval well separated from the first mode; this result does not depend strongly on the form of the tip.

It can be seen from Fig. 4 that there is a disagreement between these calculated frequencies of the IF modes and those obtained in experiment. In order to find a reason for this discrepancy we calculated the strain-induced phonon shifts. As it was established experimentally in Ref. 18 the strain in the InAs/GaAs self-assembled quantum dots has a tendency to localize near the interface being maximum at the interface and relaxing away from the interface to a barrier. Therefore, the high-index IF modes with short effective wavelengths, being localized closer to the interface than the low-index IF modes, will be more strongly subjected to the strain. Furthermore, with an increase of the excitation energy (and as a consequence with a decrease of the effective wavelength of the IF mode) all the IF modes become localized closer to the interface and thus, will be subjected to higher strain. This explains the dependence of the frequencies of the IF modes on the excitation energy (the dispersion is weaker for the more extended first mode and stronger for the highly localized high-index modes). In the limit of the long wavelengths of the IF modes their frequencies should approach the values calculated in Ref. 13 (second set of arrows in Fig. 4); this tendency is clearly observed in Fig. 4. However, at short enough wavelengths (as it was used in the experiment) the strain influences the IF modes.

We have calculated the strain-induced phonon shifts with the strain tensor obtained for the pyramidal InAs/GaAs quantum dots.[4,7] Then, the frequencies of the optical phonons in the presence of strain can be calculated as solutions of the secular equation derived in Ref. 19. The solution of the problem can be essentially simplified for the dots grown on the (001)-oriented substrates. In this case the strain at the apex of a pyramid can be described by a tensor with no shear components.[4,7] Therefore, the equation for the strain-induced LO phonon shift becomes

$$p\epsilon_{zz} + q(\epsilon_{xx} + \epsilon_{yy}) = \Omega^2 + \omega_0^2, \tag{1}$$

where $\epsilon_{xx}, \epsilon_{yy}, \epsilon_{zz}$ are the diagonal components of the strain tensor, p and q are the constants which determine the strain dependence of the phonons (they were taken from Ref. 20), and Ω and ω_0 are the strain-dependent and the bulk phonon frequencies, respectively.

The frequencies of the IF modes were calculated as follows. Near the apex of a pyramid both the strain and the electrostatic potential of the IF modes are distance-dependent.[4,6,7,13,18] The dependencies of the components of the strain tensor on the distances were taken from Refs. 4 and 7. The dependence of the amplitude of the electrostatic potential of the relevant IF modes on the distance, in the case of a discrete spectrum, is given by the power law[13]

$$f(r) \sim r^{\nu}, \tag{2}$$

where ν depends on the mode index and on the wavelength of the mode.

Then, the frequencies of the IF modes were calculated by averaging the strain-induced frequency shifts, weighted by the values of the mode potential, over the distance. The positions of the IF1 and IF2 modes, which agree well with the experimental values were calculated with $\nu = -1/3$, and they are shown in Fig. 3. It turned out that the observed strain-induced shift of the IF3 mode can be explained if a more rapid decay of the electrostatic potential than that which was used for the two first modes is considered; the frequency of the IF3 mode which better agrees with the one observed in experiment was obtained with $\nu = -1$ (the result is depicted in Fig. 3).

The comparison between the experimental and the calculated frequencies of the IF modes reflects the tendency of the IF modes to localize closer to the interface as the mode index increases (and thus, the effective wavelength of the IF mode decreases) as it was predicted by the theory.[13]

We emphasize that the observation of the strain in the GaAs barrier, surrounding the InAs dot, is itself evidence of the formation of the dots, because in the case of a thin (few monolayers thick) InAs layer embedded in GaAs, the strain should be completely localized inside the layer.

Finally, in the InAs/GaAs self-assembled quantum dots we observed the vibrational modes in the frequency range of the GaAs optical phonons. The theoretical analysis based on the calculations of the interface modes localized at sharp tips

and on the strain calculated for the pyramidal InAs/GaAs quantum dots showed that the observed modes can be assigned to the interface modes localized at the apexes of the InAs pyramids which are subjected to the strain. Moreover, we observed the strain-shifted TO and LO phonons of the InAs dot with the frequencies being in good agreement with the calculated values.

It is worth mentioning here the generality of the phenomenon observed in this investigation: although IF modes at the apexes of pyramidal dots were studied, they could be found in any corrugated interface containing tips and cusps where IF modes can be localized.

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