Raman scattering in InAs/AlGaAs quantum dot nanostructures

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We report on Raman scattering experiments on InAs/$Al_xGa_{1-x}As$ quantum dot heterostructures with $0 \leq x \leq 0.6$. The samples were prepared by using molecular beam epitaxy (MBE) and atomic layer MBE for the growth of different layers. For $x > 0$, we detected several lines originating from the $Al_xGa_{1-x}As$ alloy. These can be related to scattering from GaAs-like and AlAs-like phonons with $q \cong 0$, and weaker scattering from disorder-activated phonons with $q \neq 0$. In particular, we identified a line at ~ 250 cm^{-1} as due to disorder-activated longitudinal optical phonons in the alloy. This conclusion is different than the attribution of this line to scattering from dots and, consequently, we do not recognize the possibility of deriving any information about the actual composition of the dots from an analysis of this line as proposed by other authors. © *2011 American Institute of Physics*. [doi:10.1063/1.3567024]

In InAs/$Al_xGa_{1-x}As$ self-assembled quantum dot (SAQD) systems the increased quantum confinement with respect to InAs/GaAs QD heterostructures makes it possible, in principle, to obtain emission in the range of interest for Si-based electronics (~ 0.98 μm).[1–3] The QD characteristic emission energy depends on several parameters[4] such as confining layer (CL) composition and thickness, wetting layer composition and thickness, and QD composition and morphology. Recent works have proved the possibility of QD strain engineering to tune the characteristic emission at room temperature of InAs/GaAs QDs to wavelengths up to 1.58 μm.[4,5] In addition it has been also shown that by using a simple model,[6,7] based on a single-band effective mass approximation for QDs with cylindrical symmetry (suitable for description of the fundamental transition in QDs) satisfactory predictions for engineering the optical properties of InAs/GaAs QD nanostructures can be obtained.[4,6] On the contrary, in the case of 0.98 μm emitting InAs/AlGaAs SAQDs, discrepancies between experimental and calculated emission wavelength[8] suggest incorporation of Ga and/or Al in InAs QDs as a possible explanation for the systematic underestimation of the emission wavelength resulting from calculations based on the same model successfully applied to nanostructures emitting at longer wavelength.[4,6]

Raman scattering was proposed[9] as a method for estimating the dot composition in InAs/AlGaAs SAQDs, by means of the effect that intermixing produces on QD vibrational mode frequencies through the composition dependent strain induced in InAs dots. We planned to apply the method to InAs/$Al_xGa_{1-x}As$ SAQD systems, with the purpose of investigating the origin of the reduced QD emission blueshift. In the present letter, we present the results of Raman scattering measurements on InAs/$Al_xGa_{1-x}As$ QD nanostructures with Al mole fraction x ranging from 0.0 to 0.6.

The structures were grown on semi-insulating (100) GaAs substrates and consist of a 100 nm thick GaAs buffer layer, a 20 nm thick $Al_xGa_{1-x}As$ lower CL (LCL), the InAs QDs with 3.0 ML coverage, a 20 nm thick $Al_xGa_{1-x}As$ upper CL (UCL) with the same composition of LCL, and a 10 nm thick GaAs cap layer. Buffers and LCLs were grown by molecular beam epitaxy (MBE) at 600 °C while QDs were deposited by atomic layer MBE (ALMBE) (Ref. 10) at 460 °C; UCLs and cap layers were prepared by ALMBE at lower temperatures (360 °C) in order to reduce interdiffusion effects that may affect the QD composition and shape.[8] A reference sample with $x=0.3$ and without the QD plane was also grown in the same experimental conditions.

The spectra were taken by means of a Jobin Yvon Labram microspectrometer. The instrument was equipped with a 1800 lines/mm grating, a He–Ne laser source ($\lambda = 632.8$ nm, 15 mW), a long working distance 50× objective with 0.50 numerical aperture, and a charge-coupled device detector. The samples were placed in a flux cryostat operated with liquid N_2. The weak Raman signal in the spectral region of interest (around 250 cm^{-1}) required long integration times, typically several minutes, and maximum care in suppressing spurious signal (stray light).

The measurements were performed in $z(xy)\bar{z}$ and $z(yy)\bar{z}$ backscattering geometries. Porto notation is used; $k_i(e_ie_s)k_s$ denotes an experiment with incident light wave vector and polarization parallel to k_i and e_i, respectively, and scattered light wave vector and polarization parallel to k_s and e_s, respectively. Sample crystallographic axes were oriented parallel to laboratory axes; ($x \| [100]$, $y \| [010]$, $z \| [001]$). A mixed polarization geometry $z(yy)\bar{z}$ with $y \| [110]$, $z \| [001]$ was also used in some experiments (not presented in this letter). Sample temperature was kept at 77 K, unless otherwise stated.

Figure 1 shows a comparison between spectra obtained from several samples in $z(xy)\bar{z}$ geometry. In this experimental configuration QD-related Raman modes were observed in previous works on InAs/GaAs (Refs. 11 and 12) and InAs/$Al_xGa_{1-x}As$ QDs (Ref. 9). In our study, the dominating features in the region below 300 cm^{-1} are the GaAs LO(Γ) line at 293 cm^{-1} and the GaAs-like $LO_2(\Gamma)$ line around 280 cm^{-1}. The latter originates from the $Al_xGa_{1-x}As$ alloy of

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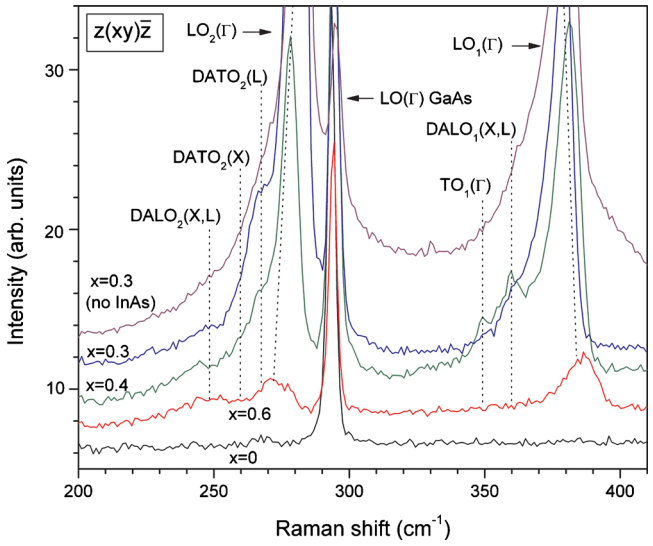

FIG. 1. (Color online) Raman spectra obtained from InAs QDs in $Al_xGa_{1-x}As$ with x=0, x=0.3, x=0.4, and x=0.6. The spectrum obtained from a sample with x=0.3 and without any InAs layer is also shown for comparison. The temperature is 77 K except for the latter sample (100 K). The symbol z(xy)$\bar{z}$ denotes the experimental configuration (see text). The spectra are vertically shifted for clarity.

CLs in samples with x>0. A band in the QD region around 250 cm^{-1} was only detected in samples with x≥0.3. No signal in the QD region could be observed in a sample of InAs dots in GaAs matrix (x=0). Increasing integration time up to 6 h was not sufficient to detect the QD signal.

Figure 2 shows spectra from the same samples obtained in z(yy)$\bar{z}$ geometry (incidentally, we notice that only spectra obtained in z(xy)$\bar{z}$ configuration were presented in the letter by Ibáñez *et al.*).[9] The GaAs LO(Γ) mode line is strongly suppressed in all samples, according to selection rules, whereas the GaAs-like $LO_2(\Gamma)$ mode does not follow the selection rule expected in bulk $Al_xGa_{1-x}As$. The feature around 250 cm^{-1} is enhanced, where detectable, with respect to z(xy)$\bar{z}$ geometry. This behavior seems incompatible with

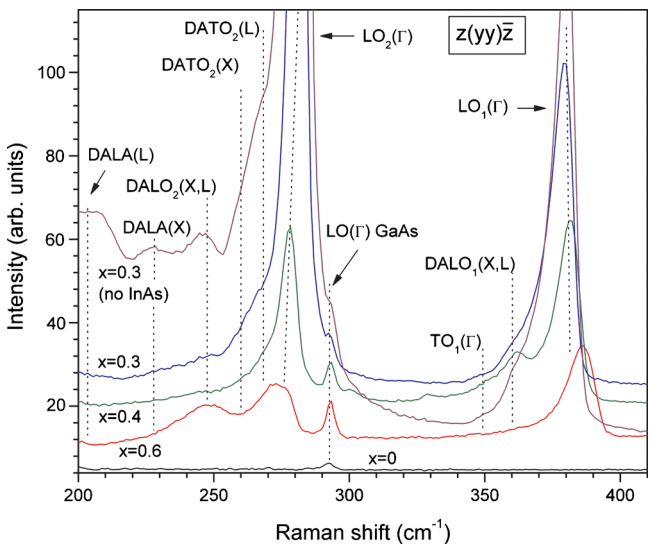

FIG. 2. (Color online) Same as Fig. 1 except for z(yy)$\bar{z}$ experimental configuration. The intensity scale is corrected for the different laser power at the sample with respect to z(xy)$\bar{z}$ configuration. The temperature is 77 K for all samples. The spectra are vertically shifted for clarity.

the attribution of this feature to the QD signal. In fact, according to previous work on InAs dots in GaAs matrix,[11] one should expect to observe InAs QD phonon lines in the z(xy)$\bar{z}$ configuration only.

Moreover, we also observed a band at 250 cm^{-1} in the reference sample with a 400 Å layer of $Al_{0.3}Ga_{0.7}As$ embedded in GaAs, i.e., a sample having the same structure of the QD sample with x=0.3 except for the absence of InAs in the structure. This band, and its intensity dependence on polarization geometry was similar to that of the samples with the QDs (see Figs. 1 and 2). This suggests that the 250 cm^{-1} feature observed in our experiments is related to the AlAs alloy, rather than the InAs dots. In fact, frequency and dependence on experimental geometry are in agreement with scattering from GaAs-like disorder-activated longitudinal optical (DALO) phonons in $Al_xGa_{1-x}As$ at X- and L-point in the Brillouin zone; $DALO_2(X,L)$.[13–17] We also identified two GaAs-like disorder-activated transverse optical (DATO) mode lines,[14,15] namely, $DATO_2(X)$ at about 260 cm^{-1}, and $DATO_2(L)$ at about 268 cm^{-1}.

The most intense line in the AlAs-like phonon region is the $LO_1(\Gamma)$ line at 380–390 cm^{-1} (see e.g., Ref. 17), which is clearly observed in the spectra of samples containing AlAs alloy (see Figs. 1 and 2). We also detected two lines between 350 and 360 cm^{-1} in the spectra of the same samples (with the possible exception of the sample with x=0.6). These lines are more intense in the z(yy)$\bar{z}$ configuration. We ascribe them to scattering from $TO_1(\Gamma)$ and $DALO_1(X,L)$ respectively.[16,17] Similar spectra were recorded in the same spectral region by Ibáñez *et al.*[9] These authors, however, only mentioned the line at higher frequency in their discussion, identifying it with $TO_1(\Gamma)$.

We observed a strong temperature dependence of spectral intensities in the samples with x=0.3 (with and without QD plane) in the range from 77 to about 120 K in all experimental configurations. In fact, the low temperature band-gap energy of $Al_{0.3}Ga_{0.7}As$ is expected to be very close to the photon energy of our laser source (1.96 eV),[18] thus allowing resonance tuning or detuning by varying the temperature. The spectral intensity at 77 K from the sample without QDs was higher than that from the sample with QD plane at the same temperature. The intensities were approximately equal if the first was heated to 100 K, which is indicative of slightly different Al mole fractions in these two samples. Measurements on the sample without QDs at 77 K (see Fig. 2) allowed to clearly detect the additional disorder-activated longitudinal acoustic (DALA) phonon lines DALA(X) and DALA(L).[14]

In summary, we studied the Raman spectra of several samples containing InAs QDs embedded in $Al_xGa_{1-x}As$. For x>0, we detected lines which are due to scattering from phonons in the $Al_xGa_{1-x}As$ alloy. Such phonons include not only GaAs-like and AlAs-like optical phonons with wave vector q≅0, according to the well known two-mode behavior, but also disorder-activated phonons with q≠0. In particular, we identified a line at ~250 cm^{-1} as due to disorder-activated longitudinal optical phonons at X- and L-point in the Brillouin zone. We discarded the alternative interpretation of this line as scattering from InAs QDs because of its intensity dependence on experimental configuration, and because we observed a line with the same characteristics even in samples containing only $Al_xGa_{1-x}As$ layers, without any

InAs layer. The present results lead us to conclude that Raman scattering from InAs QDs in $Al_xGa_{1-x}As$ around 250 cm^{-1} can hardly be observed, as a consequence of the superposition of the more intense scattering from the $Al_xGa_{1-x}As$ alloy itself. Therefore, we believe that Raman scattering from InAs dots in $Al_xGa_{1-x}As$ cannot be regarded as a practical tool to estimate QD composition, as it was suggested in Ref. 9.

[1]D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dots Heterostructures* (Wiley, New York, 1999).
[2]A. Polimeni, A. Patanè, M. Henini, L. Eaves, and P. C. Main, Phys. Rev. B **59**, 5064 (1999).
[3]F. Klopf, J. P. Reithmaier, and A. Forchel, J. Cryst. Growth **227–228**, 1151 (2001).
[4]L. Seravalli, M. Minelli, P. Frigeri, S. Franchi, G. Guizzetti, M. Patrini, T. Ciabattoni, and M. Geddo, J. Appl. Phys. **101**, 024313 (2007).
[5]L. Seravalli, P. Frigeri, G. Trevisi, and S. Franchi, Appl. Phys. Lett. **92**, 213104 (2008).
[6]L. Seravalli, M. Minelli, P. Frigeri, P. Allegri, V. Avanzini, and S. Franchi, Appl. Phys. Lett. **82**, 2341 (2003).
[7]J. Y. Marzin and G. Bastard, Solid State Commun. **92**, 437 (1994).
[8]G. Trevisi, P. Frigeri, M. Minelli, and S. Franchi, J. Electron. Mater. **37**, 937 (2008).
[9]J. Ibáñez, R. Cuscó, L. Artús, M. Henini, A. Patanè, and L. Eaves, Appl. Phys. Lett. **88**, 141905 (2006).
[10]F. Briones and A. Ruiz, J. Cryst. Growth **111**, 194 (1991).
[11]L. Artús, R. Cuscó, S. Hernández, A. Patanè, A. Polimeni, M. Henini, and L. Eaves, Appl. Phys. Lett. **77**, 3556 (2000).
[12]Y. A. Pusep, G. Zanelatto, S. W. da Silva, J. C. Galzerani, P. P. Gonzalez-Borrero, A. I. Toropov, and P. Basmaji, Phys. Rev. B **58**, R1770 (1998).
[13]D. N. Talwar, M. Vandervyver, and M. Zigone, Phys. Rev. B **23**, 1743 (1981).
[14]N. Saint-Cricq, R. Carles, J. B. Renucci, A. Zwick, and M. A. Renucci, Solid State Commun. **39**, 1137 (1981).
[15]R. Carles, A. Zwick, M. A. Renucci, and J. B. Renucci, Solid State Commun. **41**, 557 (1982).
[16]X.-j. Wang and X.-y. Zhang, Solid State Commun. **59**, 869 (1986).
[17]J. Leng, Y. Qian, P. Chen, and A. Madhukar, Solid State Commun. **69**, 311 (1989).
[18]M. El Allali, C. B. Sørensen, E. Veje, and P. Tidemand-Petersson, Phys. Rev. B **48**, 4398 (1993).