Raman scattering in InAs/AlGaAs quantum dot nanostructures

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(Received 16 February 2011; accepted 23 February 2011; published online 15 March 2011)

We report on Raman scattering experiments on InAs/Al x Ga 1−x As quantum dot heterostructures with 0 ≤ x ≤ 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 x Ga 1−x As alloy. These can be related to scattering from GaAs-like and AlAs-like phonons with q ≠ 0, and weaker scattering from disorder-activated phonons with q = 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 x Ga 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 x Ga 1−x As SAQD systems, with the purpose of investigating the origin of the reduced QD emission blue-shift. In the present letter, we present the results of Raman scattering measurements on InAs/Al x Ga 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 x Ga 1−x As lower CL (LCL), the InAs QDs with 3.0 ML coverage, a 20 nm thick Al x Ga 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. 9 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 (λ =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(001)z and z(yy)z backscattering geometries. Porto notation is used; k i (e i e i)k i 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)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(001)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 x Ga 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(Γ) line around 280 cm −1. The latter originates from the Al x Ga 1−x As alloy of

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FIG. 1. (Color online) Raman spectra obtained from InAs QDs in AlxGa1−xAs 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)Z denotes the experimental configuration (see text). The spectra are vertically shifted for clarity.

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

In summary, we studied the Raman spectra of several samples containing InAs QDs embedded in AlxGa1−xAs. For x>0, we detected lines which are due to scattering from phonons in the AlxGa1−xAs 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 acoustic (DALA) phonon lines DALA(X) and DALA(L).
InAs layer. The present results lead us to conclude that Raman scattering from InAs QDs in Al\textsubscript{x}Ga\textsubscript{1−x}As around 250 cm\textsuperscript{−1} can hardly be observed, as a consequence of the superposition of the more intense scattering from the Al\textsubscript{x}Ga\textsubscript{1−x}As alloy itself. Therefore, we believe that Raman scattering from InAs dots in Al\textsubscript{x}Ga\textsubscript{1−x}As cannot be regarded as a practical tool to estimate QD composition, as it was suggested in Ref. 9.