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Raman study of the topology of InAs/GaAs self-assembled quantum dots

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Abstract. The topology of self-assembled InAs/GaAs quantum dots was studied by resonant Raman scattering caused by the interface modes localized near the edges of the dots. Evidences were found that on both sides of the InAs layer containing the dots, their topologies show some resemblances. In addition, in the multilayered systems the evidence of the coalescence of the dots (which form vertical columns) in neighbor layers separated by the distance smaller than 25 monolayers was obtained.

The self-assembled InAs/GaAs quantum dots which are formed during the 3D overgrowth of highly mismatched materials have been extensively studied over last years because of their promising device potentialities. Nevertheless, the process of formation of the self-assembled quantum dots is not yet well understood although it evidently influences their electronic characteristics. The widely accepted point of view is that pyramidal InAs dots are formed on a thin ($\approx$ 1.5 ML thick) InAs wetting layer (see [1] and references herein). However, recent studies using cross-sectional tunneling microscopy [2, 3] and scanning transmission electron microscopy [4, 5] presented direct evidences that due to the segregation the InAs dots in forms of lenses or disks are rather embedded within the wetting layer and not on it. Moreover, as it has been shown in [4], the process of the capping of the InAs dots with GaAs changes their volume due to the redistribution of InAs from the dots to the GaAs capping layer. Obtaining information about the formation of the quantum dots in this case is very difficult. Therefore, any application of spectroscopic methods, which are simple to use, to study the topology of the self-assembled quantum dots is indispensable.

In this communication we present the Raman spectra of the InAs/GaAs self-assembled quantum dots showing that the topologies of the dots are somewhat similar on both sides of the InAs layer where they are formed. This implies in a difference between the material of the dots and the one of the wetting layer, which can be caused by a strong modification of the wetting layer between the dots due to the segregation, as it has been established in [2–5].

In order to study the topology of the InAs/GaAs quantum dots we measured the Raman scattering of the interface vibrational modes localized near the edges of the dots. The contribution of these modes to the Raman scattering, being proportional to the density of the dots, has been shown to appear at the resonance with electron excitations confined in the InAs dots [6]. Thus, the Raman lines associated with such interface modes can serve as indicators of the presence of the quantum dots.

The InAs/GaAs heterostructures containing the self-assembled InAs quantum dots were grown on (001)-oriented GaAs substrates by molecular beam epitaxy via Stranski–Krastanov growth. Structures with and without 500 Å thick cap GaAs layers were grown under the same growth conditions; the last ones were characterized by an atomic force microscope (AFM) Digital Instruments Nanoscope IIIa using the tapping mode. Samples both with a single InAs layer containing quantum dots and with multilayers separated by different GaAs spacers were investigated.

The single layer dots were grown with the nominal thicknesses of InAs 2, 2.5, and 3 ML's at the temperature $T = 500\,^{\circ}\mathrm{C}$ and the As_4 background pressure $P_{\mathrm{As}} = 8 \times 10^{-6}$ Pa. The multilayered dot structures were grown as following: after the growth of a GaAs/AlAs superlattice and a GaAs buffer, the temperature of the substrate was reduced to 450–470 °C and the InAs layer with the nominal thickness 3 ML's was deposited. The process of the formation of the dots was controlled by reflection high-energy electron diffraction (RHEED) oscillations. A transition from streaked to spotty RHEED pattern indicating a formation of the 3D islands was observed after the deposition of an effective thickness of InAs equal to 1.8 ML. The growth was interrupted for 30 s after the deposition of the nominal thickness of InAs; then the GaAs spacer of the corresponding thickness was grown and the process was repeated in order to obtain the multilayered structure. During the growth the fluxes of InAs and GaAs were fixed at 0.1 and 0.35 ML/s respectively, while $P_{\mathrm{As}} = 2 \times 10^{-6}$ Pa. Finally, the structure was capped with 500 Å of GaAs.

The Raman scattering was performed at $T = 8$ K with a Jobin-Yvon U-1000 double-grating spectrometer supplied with a conventional photon counting system. A Ti-sapphire tuned laser pumped with an Ar^{+} ion laser was used for excitation near the $E_0 + \Delta_0$ resonance of the InAs quantum dots. The cross-polarized Raman spectra were measured in order to avoid photoluminescence.

In order to obtain information about the InAs/GaAs interface we compared the Raman spectra of the samples grown with and without the GaAs capping layer. We expect that in the case of a plane InAs/GaAs interface, the interface vibrations associated with the edges of the dots would contribute to the Raman spectra in the capped samples and would not in the samples without capping layers.

As it has been shown in [6], the interface modes associated with the InAs quantum dots are seen in Raman scattering in resonance with the $E_0 + \Delta_0$ electron excitations confined in the dots ($E_{\mathrm{ex}} \approx 1.72$ eV, as measured in [7]). At such a resonance excitation the GaAs bulk phonons are week and they are detected as a shoulder at the high frequency side of the Raman line corresponding to the first interface mode [6]. The obtained Raman spectra are plotted in Fig. 1. In all the samples grown with the capping layer the Raman lines caused by the GaAs-like interface modes were observed. The first interface modes located at 293 cm^{-1} reveal larger intensities as compared to the high-index ones. Although with smaller intensities, identical lines were found in all the uncapped samples. This result testifies to the formation of the edges between the dots and the underlying GaAs (similar to those between the dots and the capping layer), which can appear due to the modification of the InAs wetting layer between the dots. As it has been shown in [2]-[5], the segregation strongly alters the contents of the wetting layer between the dots; as a consequence, the InAs dots become effectively embedded within the InGaAs wetting layer giving rise to the relevant interface modes. Actually, in this case the interface modes are localized at the edges formed by the boundary between the dots and the InGaAs wetting layer. Analyzing the ratios of the Raman line intensities measured in the uncapped quantum dots to the capped ones, we can conclude that the larger this ratio, the sharper the edges corresponding to the top of the dots relative to the edges of their bases embedded in the wetting layer.

It is worth mentioning, that the InAs-like interface modes were also found in the Raman spectra of the samples under investigation. However, due to their relatively weak intensity

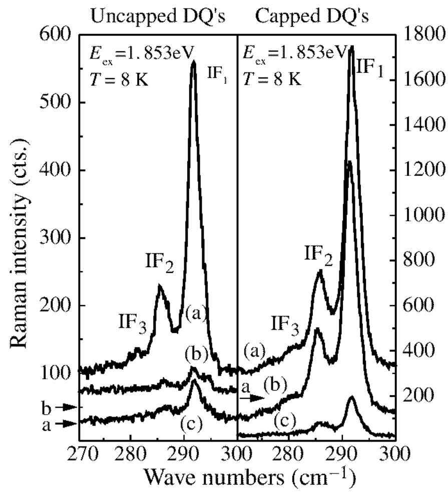

Fig. 1. The GaAs-like interface modes measured at $T = 8$ K in the single layer InAs/GaAs heterostructures containing self-assembled quantum dots grown with nominal thicknesses of InAs: (a) 2 ML's, (b) 2.5 ML's, (c) 3 ML's. The right panel shows the samples with the GaAs capping layer, while on the left one the spectra of the uncapped samples are presented. Arrows show positions of the zero intensities for the corresponding spectra, which were shifted up.

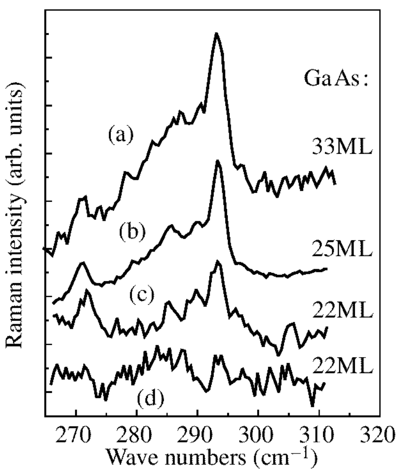

Fig. 2. The GaAs-like interface modes measured at $T = 8$ K in the multilayered InAs/GaAs heterostructures with the 2 ML's thick InAs layers separated by GaAs spacers with different thicknesses: (a) 33 ML's ($E_{ex} = 1.75$ eV), (b) 25 ML's ($E_{ex} = 1.78$ eV), (c) 22 ML's ($E_{ex} = 1.78$ eV), (d) 15 ML's ($E_{ex} = 1.77$ eV); the numbers of periods are: 15, 10, 7, and 7 respectively.

we could not include them in the analysis.

In addition, we studied the interface modes in the multilayered systems containing self-assembled quantum dots. The Raman spectra measured in the samples with different thicknesses of the spacers between the quantum dot layers are depicted in Fig. 2. As it is seen, for thicknesses of the spacers smaller than 25 ML's, the intensities of the interface modes decrease with the decrease of the spacer thicknesses. This occurs because at small thicknesses of the spacers the InAs dots in vertical columns formed by aligned growth of neighbor layers coalesce (or they are close to coalescence), thus decreasing the density of the tips responsible for the relevant interface modes. At the spacer thicknesses equal to 15 ML's a significant number of dots coalesce resulting in very weak intensities of the interface modes. Thus, this shows that Raman spectroscopy can serve as a tool to characterize the separation of the quantum dots in multilayer systems.

To summarize, by Raman spectroscopy of the interface modes localized near the edges of the InAs/GaAs self-assembled quantum dots we found an evidence of the similar topologies of the quantum dots on both sides of the layer where they are formed. We showed that the Raman scattering is a tool sensitive enough to analyze the separation between the quantum dots in multilayered systems.

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