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Under Vacuum Photoluminescence Study of Vertically Stacked InAs/GaAs Quantum Dots

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Abstract: This work presents a study of photoluminescence (PL) in vacuum from vertically stacked InAs/GaAs quantum dots on (100) N⁺ oriented GaAs substrate. A PL system in ultra-high vacuum that was coupled onto a molecular beam epitaxy (MBE) chamber was used in order to avoid the oxidation of the quantum dots. After carrying out the PL of the first layer, the same sample has been returned back to the MBE chamber to grow a second, third, forth and fifth layer of QDs, where after every layer the sample has been taken out of the MBE to the PL system. A clear double peak structure revealing a bimodal size distribution of the QDs has been shown by the power and the temperature dependent PL studies. This bimodal size distribution of the QDs has been also confirmed by an atomic force microscopy (AFM) image.

A blue shift in the PL was observed after the deposition of the second layer due to the generation of a strain field which results in the formation of a second QD layer with different structure and dimensions compared to the first QD layer.

The present observations can help improve understanding the dependence of the optical properties of InAs/GaAs QDs on inter-diffusion of In and Ga atoms during the growth of a new layer due to annealing effect.

Keywords: Photoluminescence; Vacuum; Quantum Dots; InAs / GaAs.

Introduction

Recent interest has been focused on the properties of self-organized InAs/GaAs quantum dots (QDs) due to their potential for electronic and optoelectronic device applications [1- 6]. The size uniformity of the QDs is a major problem which must be solved before high-efficiency devices can be fabricated. To achieve high performance devices, it is essential to understand the rule of thermal treatment of such devices. In view of the fact that thermal treatment may change the strain, composition and size of multiple vertically stacked InAs/GaAs QDs, it is tremendously interesting to look into the thermal effect on the optical properties and on the possibility of improving the size distributions of the QDs[7- 13]. In most cases, results were obtained on vertically stacked

InAs/GaAs self – organized QDs, by fabricating the whole stacked QDs without fully understanding the thermal effect on each layer of dots. For this purpose, a PL system in ultra-high vacuum that was coupled onto a molecular beam epitaxy (MBE) chamber was used in order to avoid the oxidation of the quantum dots due to air exposure. After carrying out the PL of the first layer, the same sample has been returned back to the MBE chamber to grow a second, third, forth and fifth layer of QDs, where after every layer the sample has been taken out of the MBE to the PL system. In this paper, I report for the first time in the literature a PL emission from each layer of the same stacked InAs QDs sample.

Experimental Details

The samples studied here were grown using a solid-source molecular beam epitaxy chamber connected to a PL system under ultra-high vacuum (UHV). A $0.3\ \mu\text{m}$ GaAs buffer layer was grown on (100) N^+ GaAs substrates at 580°C after oxide desorption. This was then followed by 2 ML of InAs and the formation of QDs at 520°C . The dots were obtained using the Stranski–Krastanov growth mode. Cycles of 0.14 ML of InAs plus a 2 s interruption under As_4 flux were repeated until the total 2 ML of InAs was deposited. The QDs were next annealed for 20 s to improve the QD size distribution. The evolution of the dots was detected using *in situ* reflection high-energy electron diffraction (RHEED). The dots were then capped with 5 nm of GaAs cap layer grown at 520°C . InAs and GaAs growth rates were set to 0.065 and 1 (ML) s^{-1} , respectively. Then, the sample has been taken outside the MBE chamber to carry out the PL measurements. After that, another layer of InAs QDs has been grown on the same sample, and the same set of PL measurements were performed on the 2 layers of QDs. This process of adding an additional layer of QDs and carrying out PL measurements under vacuum has been repeated until we obtained 5 stacked layers of InAs QDs.

PL measurements were performed in a modified Omicron variable temperature scanning

tunneling microscopy (VT-STM) chamber under UHV that was maintained to be better than 10^{-10} Torr. A set of optics inside and outside the VT-STM were used to carry out the PL measurements [14]. The VT-STM chamber was coupled to a Riber MBE system in order to avoid oxidation of the semiconductor surface. The PL measurements were performed over a temperature range of 77–300 K under excitation of a 632.8 nm line of a helium–neon laser. The luminescence was detected by a cooled InP/InGaAs photomultiplier tube (Hamamatsu R5509-73) mounted on 0.5 m monochromator. To study the surface morphology of the nanostructures, another sample having the same growth conditions with four layers was grown without a capping layer and imaged by an *ex situ* atomic force microscopy AFM. All samples were prepared and experimental tests were performed at the University of Arkansas, Fayetteville, Arkansas.

Results and Discussion

Fig. 1 shows an AFM image of the 4 stacked layers InAs/GaAs QDs. The AFM image and the size profile show a clear bimodal size distribution of the QDs in the studied samples with a typical average dot size of ~ 7 nm in height and ~ 25 nm in diameter. The average QD density is about $2 \times 10^{10}\ \text{cm}^{-2}$.

Fig. 2 shows the PL spectra under UHV at 77 K of the stacked InAs/GaAs QDs.

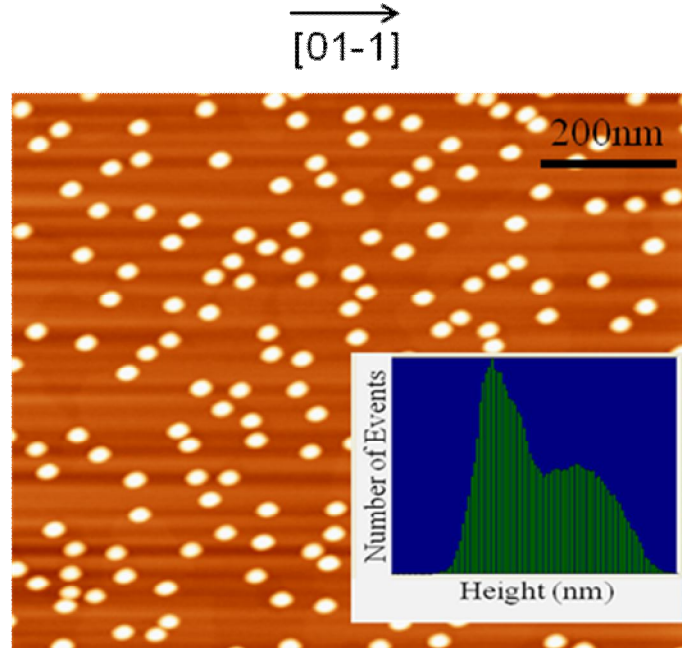


FIG. 1. ($1\ \mu\text{m} \times 1\ \mu\text{m}$) AFM image and a size profile (bottom right) of the 4 layers InAs/GaAs QDs.

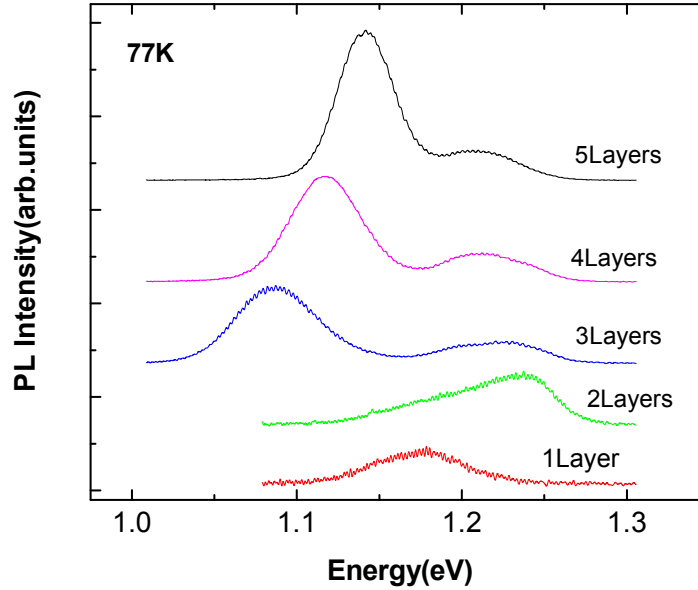


FIG. 2. The PL spectra at 77 K and under UHV of the stacked InAs/GaAs QDs.

For the first layer of QDs, one peak was observed at 1.18 eV. For closely stacked QDs, a blue shift in the PL was observed ($E = 1.24$ eV) after the deposition of the second layer. I propose that the first QD layer capped with 10 nm GaAs generates a strain field that not only causes vertical alignment but also leads to the formation of a second QD layer with different structure and dimensions compared to the first QD layer. After the deposition of the third layer of QDs, two well separated peaks were observed. The low ($E = 1.1$ eV) and the high ($E = 1.24$ eV) energy PL peaks after the deposition of the third layer are red-shifted and blue shifted, respectively, from the emission of peak of the first layer PL. These two peaks are associated with larger and smaller dots. The PL spectra after the deposition of the fourth and the fifth layers of QDs showed the same behavior as that after the deposition of the third layer. From Fig. 2, we can see that for the fourth layer, two PL peaks were observed at $E = 1.11$ eV and at $E = 1.22$ eV, and for the fifth layer, also two PL peaks were observed at $E = 1.14$ eV and at 1.22 eV. PL spectrum of the big dots clearly shifts to higher energies when the number of layers increases while the PL of the small dots does not shift.

The blue shifts of the PL spectra can be explained by inter-diffusion of In and Ga atoms during the growth of a new layer due to the annealing effect [15- 18].

In such more Ga rich dots, the transition energy increases. The Ga diffusion into small

dots stops almost after the deposition of the second layer because it approaches saturation values [15, 16].

The power-dependent PL study in Fig. 3 demonstrates that the double peaks from the 4 layers stacked QDs are associated with the ground state emission of islands in different size branches. The temperature dependence of the PL spectra from the 5 layers stacked QDs in Fig. 4 further verifies this assignment.

In Fig. 5, the PL integrated intensity of the two peaks observed in the 5 layers stacked QDs are plotted vs $1/kT$. From these plots, the thermal activation energies E_a are determined to be 240 meV and 115 meV for islands in large and small branches, respectively. Different thermal activation energies corresponding to different size islands can be measured for these samples because of the bimodal island size distribution in these samples. Two distinct island size branches coexist and can be identified by simple PL spectroscopy.

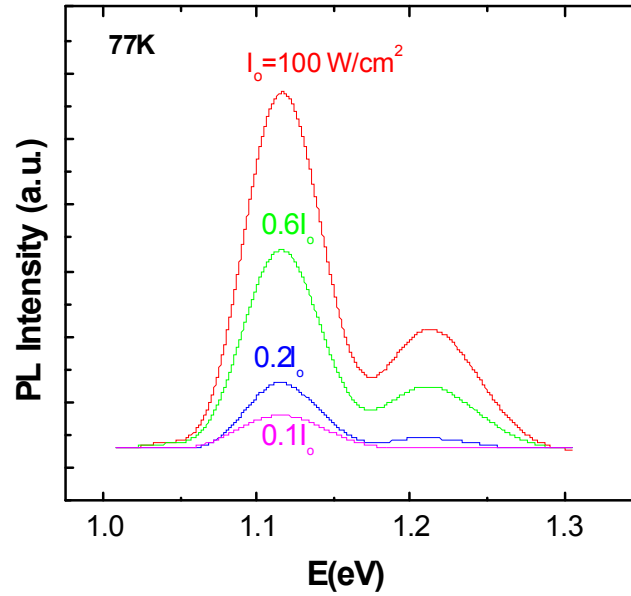


FIG. 3. The power- dependent PL spectra at 77 K and under UHV of the 4 layers stacked InAs/GaAs QDs.

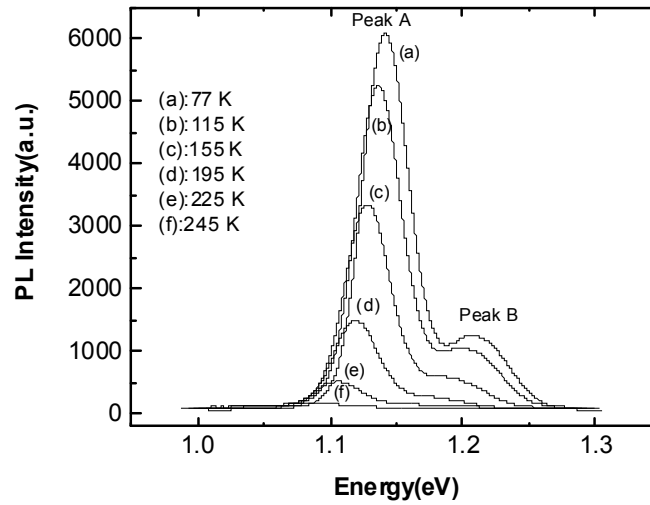


FIG. 4. The temperature dependence of the PL for the 5 layers stacked InAs/GaAs QDs.

Conclusions

The optical properties of vertically stacked InAs / GaAs QDs were investigated by using PL measurement in UHV environment. The power and the temperature dependent PL studies show a clear double peak structure, revealing a bimodel size distribution of the QDs in the studied samples. A bimodel distribution of dot sizes has also been confirmed by an AFM image. After the deposition of the second layer, a blue

shift in the PL was observed due to the generation of a strain field which results in the formation of a second QD layer with different structure and dimensions compared to the first QD layer. The blue shifts of the PL spectra for the big dots can be explained by inter-diffusion of In and Ga atoms during the growth of a new layer due to the annealing effect.

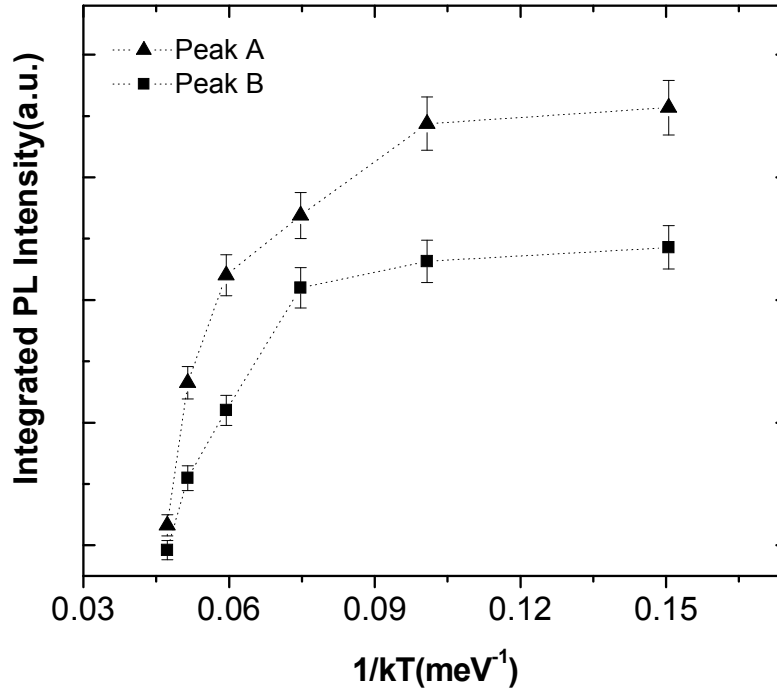


FIG. 5. Plots of PL intensities vs $1/kT$ from the ground transitions observed in the 5 layers stacked InAs/GaAs QDs.

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