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Ming Xu, Alexandre Jaffré, J Alvarez, Jean-Paul Kleider, Apichat Jittrong, et al.. Temperature dependent photoluminescence and micromapping of multiple stacks InAs quantum dots. AIP Conference Proceedings, 2015, 1649, pp.3. 10.1063/1.4913536 . hal-01239179

HAL Id: hal-01239179

<https://centralesupelec.hal.science/hal-01239179v1>

Submitted on 11 Mar 2020

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Citation: [AIP Conference Proceedings](#) **1649**, 3 (2015); doi: 10.1063/1.4913536

View online: <http://dx.doi.org/10.1063/1.4913536>

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Temperature dependent photoluminescence and micromapping of multiple stacks InAs quantum dots

Ming Xu^{1, a}, Alexandre Jaffré¹, José Alvarez¹, Jean-Paul Kleider¹, Apichat Jittrong², Thitipong Chokamnuai², Somsak Panyakeow², Mohamed Boutchich¹ and Songphol Kanjanachuchai²

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Abstract. We utilized temperature dependent photoluminescence (PL) techniques to investigate 1, 3 and 5 stack InGaAs quantum dots (QDs) grown on cross-hatch patterns. PL mapping can well reproduce the QDs distribution as AFM and position dependency of QD growth. It is possible to observe crystallographic dependent PL. The temperature dependent spectra exhibit the QDs energy distribution which reflects the size and shape. The inter-dot carrier coupling effect is observed and translated as a red shift of 120mV on the [1-10] direction peak is observed at 30K on 1 stack with regards to 3 stacks samples, which is assigned to lateral coupling.

INTRODUCTION

Semiconductors III-V QDs are crystalline nano objects that are attractive for a wide range of detection related applications particularly in QD laser¹, near infrared region (NIR)² and high efficiency solar cells³. These nanostructures are typically grown by Molecular Beam Epitaxy (MBE) using the Stranski-Krastanow (SK) growth mechanism⁴ and their small size give them interesting tunability. Indeed, the low dimensionality implies that the peak emission of QDs can be adjusted by tuning the size, composition size and passivation to suit dedicated applications. On the other hand, these objects permit the study of the transport properties of single electrons as well as advanced heterojunctions for new concepts. One major challenge remains the integration of III-V QDs on silicon substrate to develop low cost silicon photonics devices. To investigate such complex nanostructures photoluminescence has proven to be an effective tool to monitor the electronic structure as well as the optical transitions.

In this study, we employed temperature dependent macro PL characterization to investigate InAs QDs on InGaAs cross hatched patterns (CHPs) grown by MBE on (001)-GaAs substrates⁵. The structure investigated is composed of multi stack QD of 1, 3 or 5 stacks of InAs QDs and 10-nm GaAs spacer. To better understand this QDs structure, it is also important to investigate not just macroscopic PL, but also the position-dependent PL or PL mapping. For that purpose we utilized a confocal micro PL system to map the sample at room temperature and observe the crystallographic dependence of the peak emission. The temperature dependant PL spectra exhibit major differences between samples due to different QD size and lateral coupling. The PL mappings reveal position dependent luminescence⁶. These characterizations contribute to improve the understanding of the structure's optical properties, and provide a basis for future improvements on QDs based optoelectronic devices.

The Irago Conference 2014

AIP Conf. Proc. 1649, 3-7 (2015); doi: 10.1063/1.4913536
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SAMPLE PREPARATION

Quantum Dots Growth

The QDs are grown on (001) GaAs substrates with MBE. After 300nm of GaAs buffer layer, the $\text{In}_x\text{Ga}_{1-x}\text{As}$ CHP layer of 25-nm is firstly deposited on the substrate, followed by 10nm GaAs spacer layer then the QDs. Spacer and QDs deposition is repeated to form 3 and 5 stacks. At last, a 100nm capping layer of GaAs is deposited as passivation. The deposition of 10nm GaAs spacing layer and QDs is repeated to obtain 3 stacks and 5 stacks. Further details on the growth are reported in⁵. Another set of sample have been fabricated without capping to carry out AFM measurements. The final structure is shown in Fig.1 (a).

Morphology

AFM imaging demonstrates the QDs distribution on the topmost layer. As is shown in Fig.1 (b), the QDs are aligned along [1-10] and [110] dislocation lines while some of them distribute randomly for 1 stack. The diameter of QDs for the 1 stack sample is 30nm. For 3 and 5 stacks, almost all of the QDs align themselves along dislocation lines. Their morphology depends on the growth direction, the shape of the 1 stack QDs is circular but those of the 3 stacks and 5 stacks are prolonged along [1-10] direction. In contrast, the size of 3 stacks and 5 stacks are 35nm along [110] and 60nm along [1-10]. In addition, as the [1-10] QDs began to grow prior to the [110], and to free-standing ones, their sizes are a little different, that is, $[1-10] > [110] > \text{free-standing}$ ⁷, whereas the size on both directions are similar on multiple stack samples. The distance between QDs also varies with stack number. This inter-dot distance is a key parameter to favor coupling between QDs ensembles⁵. The lateral coupling rapidly decreases as the distance increases.

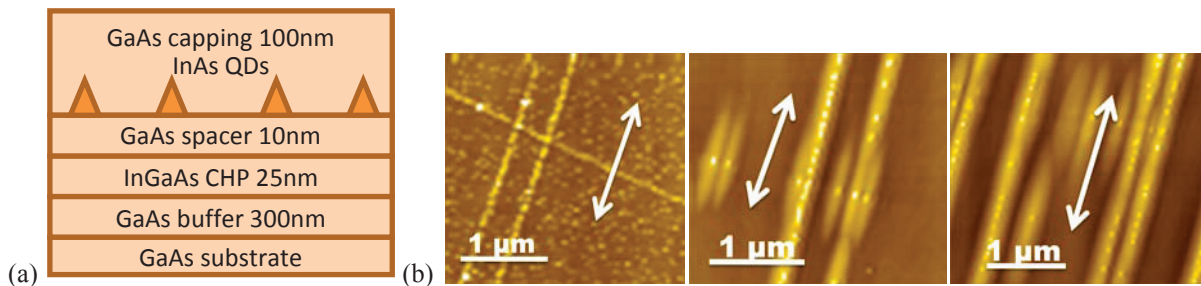


FIGURE 1. Sample structure for 1 stack sample and AFM imaging for 1, 3 and 5 stacks. Arrow in the images indicates the [1-10] direction.

TEMPERATURE DEPENDENT PHOTOLUMINESCENCE

Measurement System

Our PL system is equipped with a cryostat driven by helium circulation which allows cooling down until 20K. The excitation laser beam is projected onto the sample though cryostat window using optical fiber. The emitted luminescence is collected and focused into the monochromator and amplifier that convert photon flux to electrical signal. By controlling the monochromator, the PL spectrum is measured with a resolution of 25nm. The PL is measured under various temperatures between 30K and 300K, with a step of 10K. The sample is clamped onto the cryostat and illuminated with a 785nm laser, under a power density of 120 mW/cm². This power density permits only the ground state emissions. We performed several rounds of temperature measurements, from high to low as well as from low to high, to make sure that the measurement is stable and no hysteresis appears.

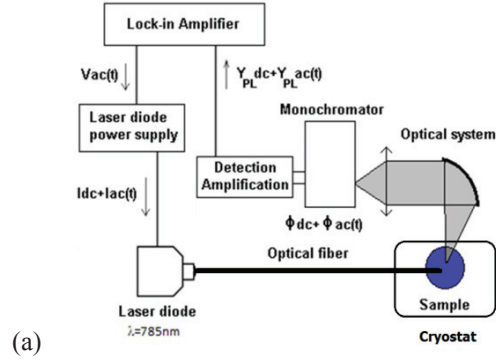


FIGURE 2. Schematic of our temperature dependent PL system

The micro PL measurements were performed with a frequency-doubled 532 nm NdYag laser operating in continuous mode as the excitation source up to 20 mW output power. The spectra were recorded through a 300 mm imaging spectrometer equipped with both 600 lines/mm and 1800 lines/mm gratings, and a back-illuminated CCD. A 100× dry and oil immersion objective with respectively a numerical aperture (NA) of 0.9 and 1.4, and a 50 μm core diameter multimode fiber acting as a pinhole were used to collect the signal. The expected lateral spatial resolution in the x-y plane can reach 150 nm (NA=1.4) and 250 nm (NA=0.9). A particular attention was paid to the incident laser power density to avoid local heating effects. A hyperspectral mapping of a 20μm by 20μm area of the sample is made possible by exciting the sample surface with 532nm laser and measuring the PL spectrum while scanning the surface using a piezoelectric stage with a spatial resolution of 10 nm.

Results

The macro PL spectra are shown in fig.2. We observe that at room temperature PL intensity is as weak as noise level, which is negligible compared to those at low temperatures. At lower temperature emission peaks begin to emerge.

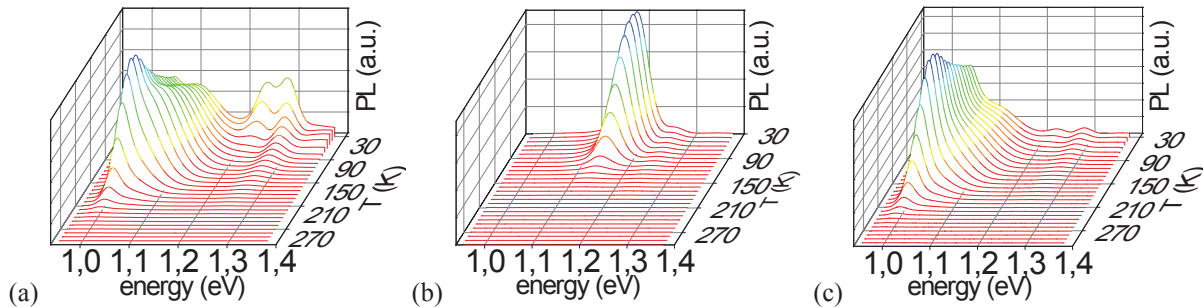


FIGURE 3. Temperature dependence of PL for (a) 1 stack (b) 3 stacks and (c) 5 stacks. The PL is measured every 10K from 30K to 300K.

Strong emissions minimize weaker ones when plotted in the same scale as is the case in Fig.3. We select some PL spectrums under various temperatures and rescaled them as shown in Fig.4 to give a clearer view of the emission peaks and the variation with temperature.

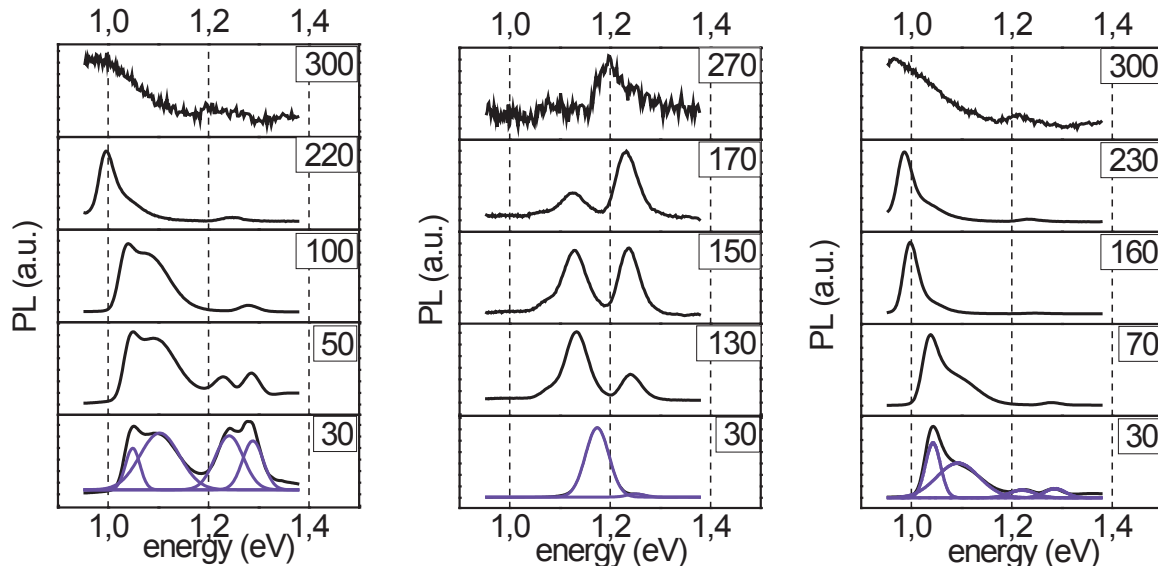


FIGURE 4. PL spectrum at some representative temperatures for 1 stack, 3 stacks and 5 stacks. Multiple Gaussian curve fitting is done for 30K to identify peak energy. 1 stack four peaks: 1.05, 1.10, 1.24, 1.29eV; 3 stacks two peaks: 1.17, 1.25eV; 5 stacks four peaks: 1.04, 1.09, 1.22, 1.28eV

In addition to temperature dependent measurement under standard PL system, we carried out micro mapping at room temperature in order to investigate the PL distribution along the various directions. Although the PL signal is relatively low at room temperature the mapping system has enough sensitivity to resolve the PL emission from noise and the images acquired confirm the QDs distribution across specific crystallographic orientations on the surface of 1 stack sample as shown in Fig.5.

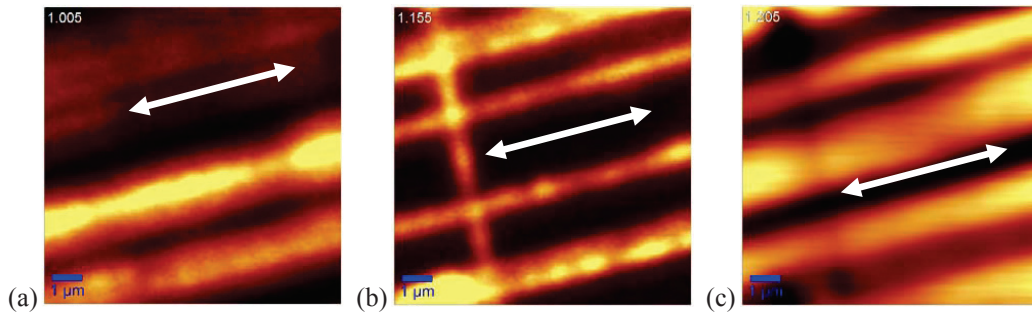


FIGURE 5. PL mapping of 1 stack at room temperature at (a) 1.005eV emission from [1-10] dots (b) 1.155eV from both [1-10] and [110] dots (c) 1.205eV from free-standing dots emission. Arrow indicates [1-10] direction.

DISCUSSION

In all samples, the emission peak of GaAs~1.42eV is not detected, indicating that the carriers generated in the GaAs and the wetting layer necessary for the growth are transferred to the QDs layer. We didn't observe vertical coupling as the spacing layer is thick enough to eliminate this effect between layers in multi-stack sample. However, strong lateral coupling is found for 1 stack.

For 1 stack, we find 4 emission peaks at 30K as is shown in Fig.4 (a). In Fig.5 the first peak is located at 1.05eV, the second at 1.10eV, the third at 1.24eV and the fourth at 1.29eV. They correspond to emission of [1-10], [110] free-standing QDs and $\text{In}_x\text{Ga}_{1-x}\text{As}$ CHP layer respectively⁷. Moreover, compared to 3 stacks, 1 stack has smaller QDs size but lower ground state energy in Fig.4.(a)-(b). This is characterized by a red shift of 120mV on the [1-10] direction peak is observed at 30K when comparing 1 stack to 3 stacks samples. This shift is assigned to lateral coupling as discussed in⁸ when the dots are very close to each other as indicated in AFM imaging.

Due to lack of free-standing QDs, the 3 stacks and 5 stacks show little of the corresponding emission. The 3-stack spectrum has two peaks and when temperature increases they both decrease as a result of thermal quenching. The intensity of the first broad peak decreases much more rapidly than the second one, making the second peak only visible at relatively higher temperature as is shown in Fig.5 (b). At 30K, the first peak is at 1.17eV and the second at 1.25eV. In fact the first peak is comprised of two closely located peaks that correspond to [1-10] and [110] dots respectively which are very close in size and shape as discussed above about Fig1.(b). The distances between QDs are bigger than 1stack, far enough not to induce coupling effect.

The behavior of 5 stacks is not similar to that of 3 stack in terms of spectra although their AFM morphology in Fig.1(b) quite resemble to each other. Inhomogeneity caused during the sample growth may be responsible for this discrepancy.

Due to the detection sensitivity, we get a noisy measurement result of 1stack at room temperature as is shown in Fig.4(a). There is a wide peak at about 1.0eV and a small one at 1.2eV. However, micro-mapping in Fig.5 demonstrates that 1.0eV emission comes from [1-10] QDs and that although we cannot see in macro-PL spectrum, [110] QDs are contributing to 1.155eV emission as shown in Fig.5(b). The 1.2eV emission should be attributed to free-standing QDs by comparing Fig.5(c) to (a) and (b).

CONCLUSION

We observed the temperature dependence of photoluminescence of multiple stack InGaAs quantum dots fabricated with MBE. The emission peaks are characteristic of the various QDs patterns present in the multi stack. In addition, using micro PL mapping, it is possible to resolve the emission peak along the crystallographic orientations and therefore identify the different QDs patterns. A blue shift of 120meV is observed on the [1-10] direction peak at 30K between the 3 and 5 stacks sample, this is assigned to the lateral coupling between dots.

ACKNOWLEDGMENTS

Authors wish to thank the French Ministère des affaires étrangères et européennes (MAEE) and the Centre National de la Recherche Scientifique (CNRS) for their financial support in the STIC ASIA program, and Thailand Research Fund (RSA5580015), as well as China Scholarship Council (CSC).

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