

GROWTH, OPTICAL AND STRUCTURAL CHARACTERIZATION OF InP NANOSTRUCTURES WITH $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ INSERTION LAYER

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Abstract

Quantum dots in InP/GaAs system were grown by low pressure MOVPE via the Stranski-Krastanow growth mode. In order to control the dots diameter and improve the size uniformity and photoluminescence (PL) emission, the ternary $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ layers thickness (0-4) monolayers (MLs) were inserted. The growth of thin $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers between $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ matrix and InP QD layers reduced the mean height and size fluctuation and significantly improved the uniformity of InP QDs. A higher QDs PL intensity, better uniformity and smaller QDs size had been achieved at 2 ML thickness InGaP insertion layer. The PL emission could be observed around 780 nm red spectral range. There was no blue-shift with increase of the $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layer thickness.

Keywords: InP, InGaP, Self-assembled Quantum Dots (SAQDs), Metal-Organic Vapor Phase Epitaxy (MOVPE), Atomic Force Microscopy (AFM), Photoluminescence (PL)

Introduction

Self-organization during growth is the prevalent method for the formation of quantum dots (QDs), structures which are not only inherently important for the understanding of quasi-one-dimensional quantum systems, but they also offer promising improvements in optical devices [1]. One of the approaches that used to achieve single-photon generation is based on the emission of semiconductor QDs [2]. In the future, one can think of a simple QD device for using in computer or networking applications. For these purposes, optically or electrically addressable single QDs are needed on a mass production scale which favors metal-organic vapor-phase epitaxy (MOVPE) due to several advantages [3,4]. Current silicon based single-photon detectors have their highest photon detection efficiency in the red spectral range, therefore it is preferable to fabricate single QD emitting at such wavelengths [5]. By using InP QDs embedded in $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ emission in this spectral range can be achieved [6]. However, InP/InGaP SAQDs on GaAs are usually formed with poor size uniformity compared to that of InAs/GaAs QDs [7,8]. Especially, inhomogeneous broadening in optical spectra due to the randomness in the dot size has been a difficult issue of limiting potential benefits. While in the case MOVPE of InP/InGaP SAQDs, a bimodal size distribution for the coherent islands or dots has often been observed at low coverages of InP [9]. This bimodal size distribution can be overcome by the insertion of GaP and InGaP insertion layers [10]. Nevertheless, the island size still remains large and hence the areal density is low [11,12]. Since the large dots may

introduce misfit dislocations and low areal density of dots gives poor optoelectronic efficiency, growth of small size, high density and highly uniform InP/InGaP SAQDs becomes imperative [13].

In this paper, we present the main experimental evidence of InP QDs embedded in InGaP matrices with insertion of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ layers grown on GaAs (100) substrates by Metal-Organic Vapor Phase Epitaxy (MOVPE) via the Stranski-Krastanow (S-K) growth mode. The QD (or island) densities $\sim 10^9 \text{ cm}^{-2}$ and size distribution and optical properties of InP QDs have been reported. In the theoretical model of the S-K growth mode, QD properties depend both on the strain and the surface condition of the layer upon which the dots are grown. Therefore, the insertion of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ interface layers between $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ matrix and InP QDs layer are also expected to change the morphology, growth characteristics and optical properties of the InP SAQDs.

Experimental Details

The growth of InP embedded in $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ matrix were carried out in a horizontal MOVPE reactor AIXTRON, model AIX200/4 with a rotating substrate holder on nominally (001) oriented GaAs substrate. The inlet of the reactor is divided into two parts: Group-III precursors were introduced from the upper inlet and group-V precursors were introduced from the lower inlet. Hydrogen gas was used as the carrier gas for precursors and as coolant between the inner reactor and the outer tube. The reactions occur in a rectangular inner liner tube, which has a graphite rotator as a sample susceptor. During MOVPE growth, GaAs substrates were placed at the center of the susceptor. For InP QDs on GaAs substrate growth, trimethylgallium (TMGa) and trimethylindium (TMIn), tertiarybutylarsine (TBAs) and tertiarybutylphosphine (TBP) were used as source precursors.

Epitaxial growth conditions were a total pressure of 100 mbar, H_2 total flow rate of 13,000 sccm (sccm denotes cubic centimeter per minute at standard pressure), at 610 °C, and V/III ratio of source precursors of 18 for InP. Lattice-matched $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}/\text{GaAs}$ structures are becoming major III-V semiconductor systems. Because, they have lower reactivity with oxygen, and more reduced DX centers and lower interfacial recombination rates, compared to AlGaAs/GaAs systems. The fabrication of InP SAQDs in InGaP/GaAs systems is difficult by metal organic vapor phase epitaxy (MOVPE), mainly due to the exchange or intermixing between As and P. The other causes that contribute to the difficulty include the ordering effect of InGaP and the segregation of In in the InGaP layer.

Schematic representation of the InP QDs embedded in InGaP matrix grown on (001) GaAs substrate was depicted in figure 1. A 120 nm GaAs buffer layer was grown on semi-insulating GaAs (001) substrates at 610 °C. After the growth of GaAs buffer, the growth of 150 nm lattice-matched $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ layers was followed at the same temperature. In all growth process, the growth temperature was fixed at 610 °C. Then, a $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layer (IL) with a varying thickness (0 - 4 MLs) was deposited to improve QDs size uniformity. After that 2 ML GaP layer was grown to get better QDs quality. Finally, the single-layer of self-assembled InP QDs was grown at a growth rate of 0.5 ML/s by depositing 4 ML of InP. After the growth of InP QDs, 50 nm cap of InGaP followed in the case of samples planned for PL measurements. The surface morphological and optical properties of InP QDs due to insertion of 0 – 4 MLs GaP and $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers were characterized by atomic force microscopy (AFM) and photoluminescence (PL). The AFM measurements were performed by using a nanoscope in close-contact mode. PL measurement was carried out using the 532 nm line of solid state laser. The PL signal was collected by an InGaAs photo-detector with solid-state laser.

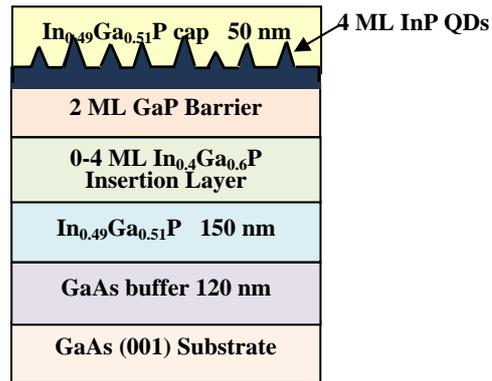


Figure 1. Schematic diagram of the vertical layer structure of InP QDs grown on (001) GaAs substrate by insertion of InGaP layer.

Results and Discussion

Structure Properties of InP Quantum Dots

The AFM images of the InP QDs grown by insertion of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ layers are shown in figure 2. The average height and diameter of InP QDs without $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL are 17 nm and 70 nm. Generally, both size and height are generally decrease by increasing the thickness of GaP insertion layer. The sample with 2 ML GaP insertion layer showed a significantly improved size, height dispersion and homogeneity. The dot density without $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL is $4.2 \times 10^9 \text{ cm}^{-2}$. After insertion of 1 ML $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL, QD density changes to $3.1 \times 10^9 \text{ cm}^{-2}$ and then slightly increases again to $3.3 \times 10^9 \text{ cm}^{-2}$ while increasing the thickness of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL to 2 ML. After insertion of 2 ML $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL thickness, the QD size was quite increased and density was also slightly increased again. This observation indicated that the thickness of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL did not significantly increase the density of QDs. It is likely that the incorporation efficiency of In during the deposition of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL layer reduces as the strain increases. The smaller and better uniformity of InP QDs at 2 ML $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL is around 16 nm height and 50 nm diameter.

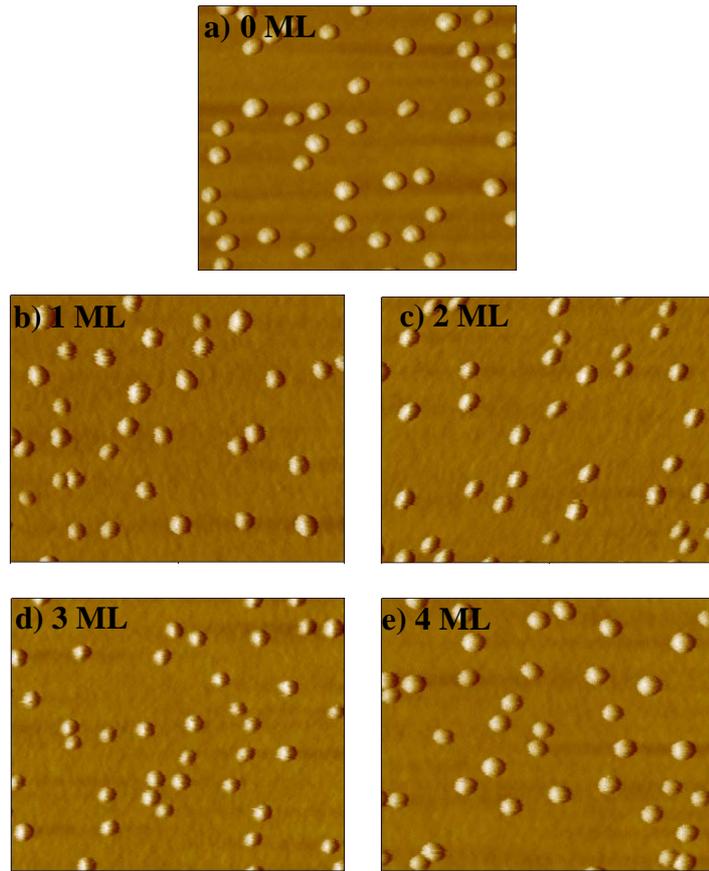


Figure 2. Typical ($1\mu\text{m} \times 1\mu\text{m}$) scan range AFM images of InP QDs embedded in InGaP barrier with $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers (0-4) MLs.

A comparison of density and diameter of InP QDs with $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers are shown in figure 3. By using an InGaP IL, the average height and diameter of InP QDs are reduced to 16 nm and 50 nm and these are also good for QDs quality [14]. The introduced strain in the lower $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ barrier strongly influences the InP QD growth, in a sense that the same amount of material is deposited but is rearranged in more and smaller QDs. This behavior becomes also obvious in the QD density, compared to the case without $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL (highest InP QD density: $4.2 \times 10^9 \text{ cm}^{-2}$) the density is reduced to $3.3 \times 10^9 \text{ cm}^{-2}$. Since the QDs growth conditions are the same, the smaller QD size and lower density for the samples grown with InGaP IL results in less incorporation of the material. On the InGaP surface, there could be an indium segregated layer which may be favorable for the nucleation of InP QDs leading to increased QDs density. The $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ insertion layer may consume this segregated indium layer thereby block preferential nucleation sites. As a result, besides the QDs density slightly increase and mean QDs height, the QDs size fluctuation is also increases. By increasing of $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ insertion layer thickness, InP QDs density increases, not too much changes of size and QDs uniformity is improved. This is also a result of the influence of $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ IL on the topmost layer InP QDs.

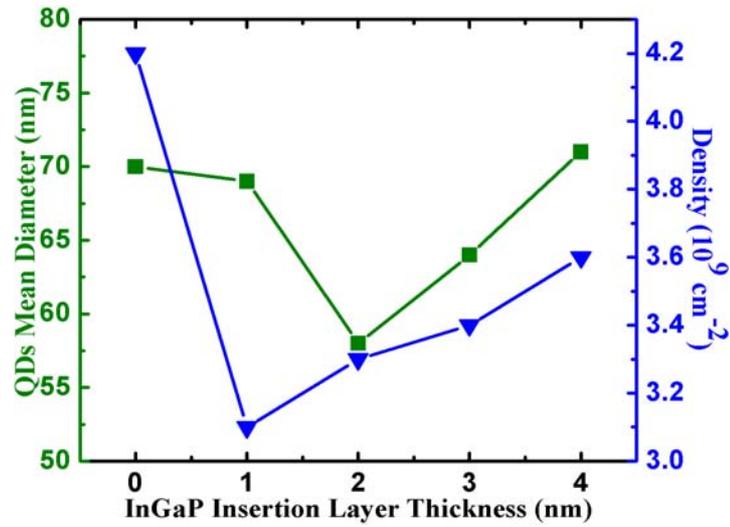


Figure 3. Effect of $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ insertion layers on InP SAQDs embedded in InGaP grown at 610 °C.

Optical Properties of InP Quantum Dots

The room temperature (RT) PL spectra of InP QDs with $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers are shown in figure 4. The ensemble PL measurements reveal already drastically changed optical properties of the InP QDs grown with varying $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL thickness. The PL spectrum of the sample without $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL shows PL peak at 781 nm. When the InGaP layer is inserted between the GaP layer and the underneath InGaP layer, a significant increase in PL intensity is observed while the line-width of the spectrum remained almost unchanged from the PL peak without $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL. Since the less QDs size fluctuation of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL samples, the better PL line-width as can be seen from the PL spectra. Since the less QD size fluctuation of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL samples, the better PL line-width as can be seen from the PL spectra. In sample with $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL, 2 ML thickness is noticeably improved intensity among other thicknesses. It indicates that an increased number of optically active InP QDs at this layer thickness. In the PL spectra of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL samples, the PL emission of InP QDs is at 777 nm. This red spectral range is also preferable to generate highest photon detection efficiency for single-photon detectors. Additionally, the InP QDs with $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ ILs must influence the optical properties of possible quantum optic devices which have to be carried out in future work.

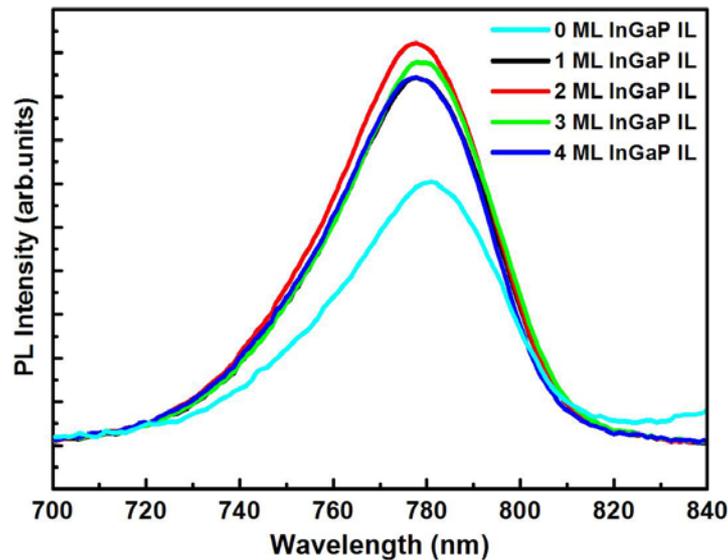


Figure 4. The room temperature PL spectra of the InP QDs grown on the $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ barrier with 0- 4 ML thick $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers.

Conclusions

We have presented an approach of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers effect on the structural and optical properties of InP QDs grown by MOVPE. The insertion of 0- 4 ML $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ layers achieves slightly increase density and it also reduces the size and height of QDs that were the better conditions for InP QDs. Thin $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ insertion layers effect on InP QDs led to improve intensity of the PL peak. As a result of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ IL samples, besides the increase of QDs density and mean diameter, the QDs size fluctuation also decreases and thus the broad of PL linewidth reduces and PL intensity increases. Under the same growth conditions, 2 ML ILs thickness is the optimum where QDs mean size and fluctuation are minimum while giving the higher PL intensity than other thickness of $\text{In}_{0.4}\text{Ga}_{0.6}\text{P}$ ILs. Since InGaP ILs improve the structure and PL quality of the InP QDs.

References

- [1] Y. Arakawa, H. Sakaki, *Appl. Phys. Lett.* Vol. 40, pp. 939, 1982.
- [2] S. P. DenBaars, C. M. Reaves, V. Bressler-Hill, S. Varma, W.H. Weinberg, and P. M. Petroff, *J. Cryst. Growth* Vol. 145, pp. 721, 1994.
- [3] W. T. Tsang, *J. Cryst. Growth* Vol. 120, pp. 1, 1992.
- [4] A. G. Thompson, R. A. Stall, W. Kroll, E. Armour, C. Beckham, P. Zawadzki, L. Aina, and K. Siepel, *J. Cryst. Growth* Vol. 170, pp. 92, 1997.
- [5] D. Richter, R. Roßbach, W.-M. Schulz, E. Koroknay, C. Kessler, M. Jetter, and P. Michler, *Appl.Phys.Lett.* Vol. 97, pp. 63107, 2010.
- [6] V. Zwiller, H. Blom, P. Jonsson, N. Panev, S. Jeppesen, T. Tsegaye, E. Goobar, M.-E.Pistol, L. Samuelson, and G. Björk, *Appl.Phys.Lett.* Vol. 78, pp. 2476, 2001.
- [7] H.-W. Ren, K. Nishi, S. Sugou, M. Sugisaki and Y. Masumoto: *Jpn. J. Appl. Phys.* Vol. 36, pp. 4118, 1997.
- [8] D. Leonard, K. Pond and P. M. Petroff: *Phys. Rev. B* Vol. 50, pp.11687, 1994.
- [9] S. P. DenBaars, C. M. Reaves, V. Bressler-Hill, M. Krishnamurthy and W.H. Weinberg: *J. Cryst. Growth* Vol. 145, pp. 721, 1994.

- [10] N. Carlsson, K. Georgsson, L. Montelius, L. Samuelson, W. Seifert and R. Wallenberg: *J. Cryst. Growth* Vol. 156, pp. 23, 1995.
- [11] W. Seifert, N. Carlsson, J. Johansson, M. E. Pistol and L. Samuelson: *J. Cryst. Growth* Vol. 170, pp. 39, 1997.
- [12] P. M. Petroff and S. P. DenBaars: *Superlattices & Micristruct.* Vol. 15, pp.15, 1994.
- [13] H-W Ren, M. Sugisaki, J-S Lee, S. Sugou and Y. Masumoto, *Jpn. J. Appl. Phys.* Vol. 38, pp. 507, 1999.
- [14] W.-M. Schulz, R. Roßbach, M. Reischle, G. J. Beirne, M. Bommer, M. Jetter, and P. Michler, *Phys. Rev. B* Vol. 79, pp. 35329, 2009.