In-mole-fraction of InGaAs Insertion Layers Effects on the Structural and Optical Properties of GaSb Quantum Dots Grown on (001) GaAs Substrate

Suwit Kiravittaya[1], **Kamonchanok Khoklang**[2], **Supachok Thainoi**[3], **Somsak Panyakeow**[4], and **Somchai Ratanathammaphan**[5], Non-members

ABSTRACT

GaSb quantum dots (QDs) have been grown by solid-source molecular beam epitaxy on a 4-monolayer (ML) $In_xGa_{1-x}As$ (x = 0.07, 0.15, 0.20, and 0.25) to investigate the effects of In-mole-fraction of InGaAs insertion layers on the structural and optical properties of the GaSb QDs. The density of grown GaSb QDs is approximately 1.2-2.8$\times 10^9$ cm^{-2} on InGaAs insertion layers which depends on the In-mole-fraction. Dot shape and size change substantially when In-mole-fraction of InGaAs insertion layers is varied. The uniformity of GaSb QDs improves when the indium content increases. The change in free-standing QD morphology is likely due to the modified strain at different values of indium compositions in InGaAs insertion layers. The effects of In-mole-fraction of InGaAs insertion layer on optical properties of the QDs are studied by photoluminescence (PL). PL results show the blueshift of the emission when the indium content in InGaAs insertion layer increases.

Keywords: InGaAs Insertion Layers, GaSb Quantum Dots, Molecular-beam Epitaxy System, GaAs, Stranski-Krastanov

1. INTRODUCTION

Semiconductor quantum dots (QDs) have gained considerable interests due to their promising properties for novel device applications in the past few decades. Many electronic and optoelectronic devices based on IIIV QDs have been realized and investigated [1-6]. They include single-electron transistor, QD laser, light-emitting diode (LED), photodectectors and memory devices. Among these devices, self-assembled In(Ga)As/GaAs QDs grown in Stranski-Krastanov growth mode are major investigated. However, spontaneous formation of QDs on substrate surface allows us to fabricate not only In(Ga)As/GaAs QDs but also QDs in many different strained material systems. GaSb/GaAs QDs, which have a type-II band alignment, might be useful and more suitable for some applications. For example, the GaSb/GaAs QDs have been proposed to use in memory device application. Due to a very large valence band offset of $\sim$0.6 eV, long storage time ($> 10^{-5}$ s) is expected [5]. Longer storage time might be archived by using GaSb QDs grown on different materials such as AlAs.

Challenges in QD technology are related to the control of spontaneous QD formation process [7]. In order to utilize QD technology, detail understandings of natural growth process must be first investigated. For most of QD applications, these self-assembled QDs must be capped. The segregation and intermixing effects for each material system e.g. GaSb/GaAs must be understood. The performance of QD devices depends on the electronic properties. Probing them by structural and optical investigations is necessary.

For optoelectronic devices, GaSb/GaAs QDs have been realized and some unique properties on different devices, such as wide tunable wavelength for LED and higher operation temperature for infrared photodetectors due to the large hole confinement and long carrier lifetime, have been demonstrated [8]. Device performance strongly depends on the QD properties. It is therefore of interest to investigate structural properties of obtained QDs under various growth conditions in order to fine-tune them for any specific applications. Nevertheless, the studies on the structural and optical properties of GaSb/InGaAs/GaAs QDs are still lacking.

In the past, there are reports on the growth as well as structural and optical properties of InAs QDs grown in InGaAs matrix [9,10]. This structure is known as dot-ina- well structure. The obtained InAs QDs have significant different properties. Due to the increasing number of adjustable parameters, perfor-

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[1] The author is with Advanced Optical Technology (AOT) Laboratory, Department of Electrical and Computer Engineering, Naresuan University, Phitsanulok, Thailand, E-mail: suwitki@gmail.com
[2,3,4,5] The authors are with The Semiconductor Device Research Laboratory (SDRL), Department of Electrical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok, Thailand, E-mail: kamonchanok.r19@gmail.com, supachok.t@chula.ac.th, s_panyakeow@yahoo.com and Somchai.R@chula.ac.th

mance of specific devices based on this structure can be optimized. In this work, we are interested in tuning the GaSb QD properties by adjusting the indium content of InGaAs layer below GaSb QDs.

In this paper, we present a study on the structural and optical properties of GaSb/InGaAs type-II QD material system with $In_xGa_{1-x}As$ insertion layers with $x = 0.07$, 0.15, 0.20, and 0.25 are introduced. All samples are fabricated by molecular beam epitaxy (MBE) and the QD structures are investigated by atomic force microscope (AFM). The photoluminescence (PL) measurements are performed to examine the optical properties of the samples at 20 K by lock-in method using a 514.5-nm Ar ion laser as an excitation source and a liquid-nitrogen cooled InGaAs photodetector.

2. EXPERIMENTS

Samples studied here were grown on (001) GaAs substrates in a solid-source molecular beam epitaxy system (Riber Compact 21) equipped with a reflection high-energy electron diffraction (RHEED) observation system and an Sb valved cracker cell. Other cells (Ga, In and As_4) are standard effusion cells. Valved cracker cell is used in this work because a relatively fast switching time between As-rich and Sb-rich environment is needed. After the desorption of surface oxide on GaAs substrate at 580°C under As4 beam in As-rich condition, a 300-nm thick GaAs buffer layer was grown at 600°C with the growth rate of 0.5 monolayer per second (ML/s). During the growth, RHEED pattern was observed and well-prepared GaAs buffer layer (flat (001) GaAs surface) showed a clear (2×4) surface reconstruction. In order to grow InGaAs insertion layer, the substrate temperature was ramped down to 500°C. After the substrate

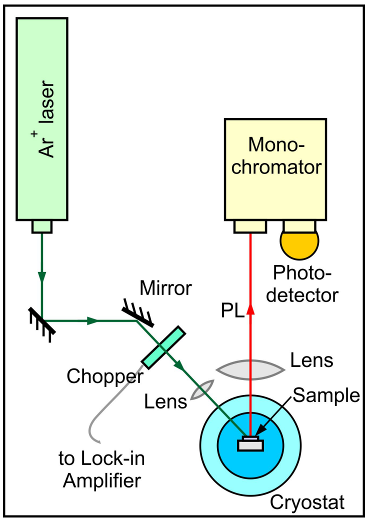

Fig.1: *Schematic of the PL setup for optical investigation of GaSb QD samples.*

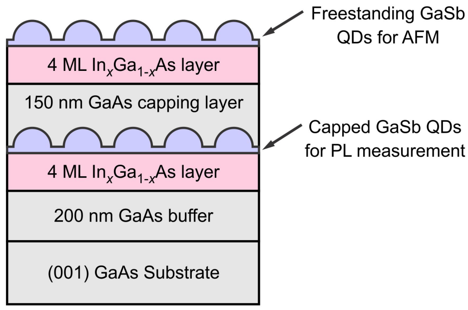

Fig.2: *Schematic structure of 3-ML GaSb QDs with 4-ML InGaAs insertion layer on undoped (001) GaAs substrate.*

temperature stabilized, 4-ML $In_xGa_{1-x}As$ was grown. Indium and gallium growth rates were changed in order to obtain the desired content of indium in InGaAs layer. For $x = 0.07$, 0.15, 0.20, and 0.25, the indium (gallium) rate are 0.025, 0.025, 0.025, and 0.033 (0.33, 0.14, 0.10, and 0.10) ML/s, respectively. After the 4-ML InGaAs insertion layer growth, the sample surface was soaked by Sb flux for 60 s. Self-assembled GaSb QDs are obtained by depositing 3-ML GaSb at the gallium growth rate of 0.1 ML/s. The V/III flux ratio (Sb/Ga) is kept constant at 4. For optical investigation, the QDs were capped with a 150-nm-thick GaAs and the 4-ML InGaAs insertion layer and GaSb QDs were grown again on top surface without capping. The top GaSb QDs are for surface morphology examination. After the growth, the sample was cooled down immediately. The surface morphology was characterized by AFM (Seiko SPA-400) in dynamic force mode in air. For PL tests, a 514.5-nm Ar-ion laser was used an excitation source, and data were obtained by a monochromator (HORIBA Jobin Yvon, iHR320) and InGaAs photodetector (Hamamatsu G9494). During the PL measurement, the sample is cooled down to 20 K by using a closed circuit helium cryostat. The schematic of the PL setup is shown in Fig. 2 and the schematic structure of the investigated samples is displayed in Fig. 2. For this work, only the indium content of InGaAs insertion layer is varied.

3. RESULTS AND DISCUSSION

Figure 3 shows AFM images of GaSb QDs on $In_xGa_{1-x}As$ (Figs. 3(a)-(d)) surfaces from the central regions of each sample. On the left column, AFM images with the colorscale corresponding to the height scale are shown while the same images with the colorscale corresponding to the slope scale is shown in the right column. The slope scale is obtained by calculating the amplitude of the surface gradient, i.e., if we define the AFM height data as $h(x, y)$, the image with the colorscale corresponding to the surface slope is

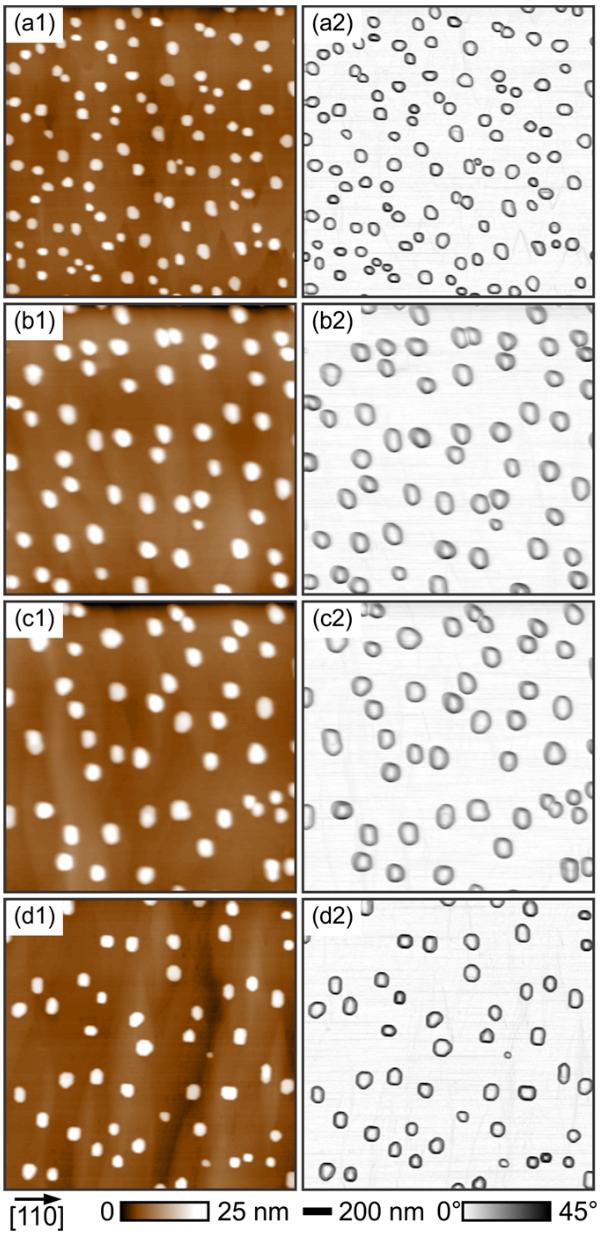

Fig.3: *(a) - (d) 2×2 μm² AFM images of GaSb QDs on 4-ML $In_xGa_{1-x}As$ with x = 0.07, 0.15, 0.20, and 0.25, respectively. Left column shows the images in height scale and right column shows the images in slope scale.*

$$|\nabla h| = \sqrt{\left(\frac{\partial h}{\partial x}\right)^2 + \left(\frac{\partial h}{\partial y}\right)^2}. \quad (1)$$

From all AFM images, we can clearly indicate the GaSb QDs on the surface. Wetting layer surface has a natural pattern of surface steps aligned along [1-10] direction. GaSb QDs are typically located on the steps and well-distributed over the whole surface. This result is consistent with the reports of the InAs/GaAs QD growth [11].

Distinct surface morphology is observed when InGaAs layer is introduced. The dot densities are approximately 1.2-2.8×10⁹ cm⁻². QD density substantially decreases and QD size increases when the indium content increases from $x = 0.07$ (Fig. 3(a)) to 0.25 (Fig. 3(d)). Gradual morphology changes are still observed. The elongation direction of the base slightly changes from [110] to [1-10] when InGaAs insertion layer is introduced. This is due to the presence of different amount of indium in the InGaAs insertion layer. The uniformity of GaSb QDs improves when the indium content increases. Increasing more indium content into InGaAs insertion layer might induce self-assembled InGaAs QD formation prior to the growth of GaSb QDs.

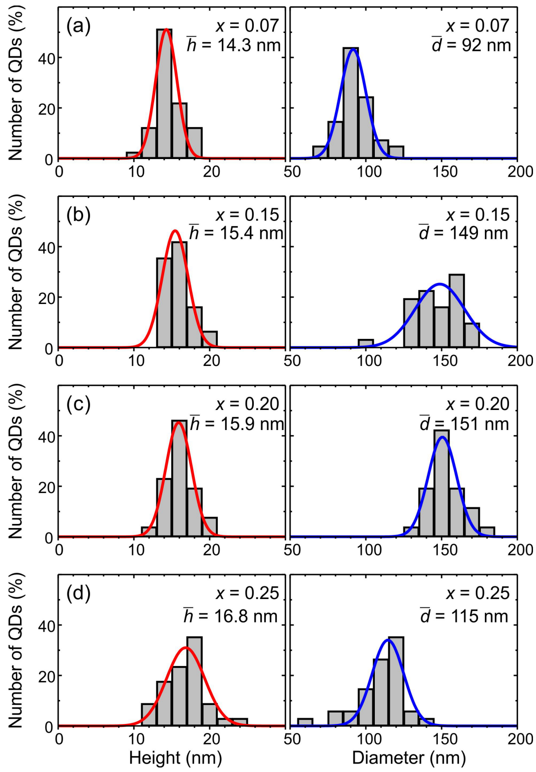

Fig.4: *(a) - (d) histograms of QD height and diameter distribution of GaSb QDs on 4 ML $In_xGa_{1-x}As$ with x = 0.07, 0.15, 0.20, and 0.25, respectively. Solid lines are Gaussian fits. Average height $\bar{h}$ and average diameter $\bar{d}$ are indicated in the figures.*

From each AFM image of GaSb QDs shown in Fig. 3, analysis of height and diameter of each freestanding GaSb QD is performed. QD height is defined as the difference between surface level of QD apex and its base while the diameter is calculated from the largest closedloop contour line of individual QD. Area is converted to QD diameter d by using circular approximation, i.e.,

$$d = \sqrt{4A_c/\pi}, . \quad (2)$$

where A_c is area of the largest closed-loop contour line. Height and diameter distribution of GaSb QDs

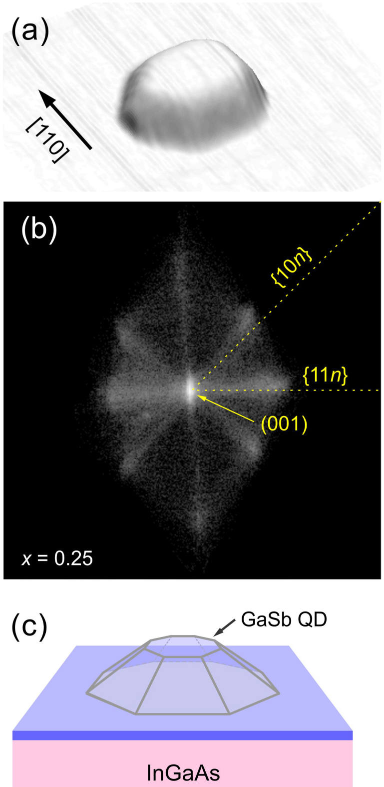

Fig.5: *(a) a magnified AFM image of single GaSb QD grown on 4-ML $In_{0.25}Ga_{0.75}As$, (b) facet plot of the free-standing QD surface and (c) schematic of the GaSb QD shape grown on InGaAs.*

on different InGaAs insertion layers is displayed as histograms in Fig. 4. All height and diameter data can well be fitted with Gaussian (normal) distribution functions. From this result, one can clearly see that both QD height and diameter increase when the diameter increase, except only at the indium content of $x = 0.25$ where we observe the reduction of diameter. This non-monotonic diameter change might be due to the excessively high indium content on the surface. Some indium atoms may diffuse and incorporate into the GaSb QDs. The presence of indium in GaSb increases the mismatch strain and therefore modifies the QD shape. These observations are due to the change of initial surface energy, interface energy as well as the strain energy of the QD system [12].

Closer look on individual QDs, one can observe distinct QD shape [13]. For GaSb grown on InGaAs insertion layer, the free-standing QDs have a flat (001) surface on the top. Figure 5(a) shows a magnified AFM image under perspective view. The GaSb QD is grown on $In_{0.25}Ga_{0.75}As$ surface. In order to quantify the orientation of QD surface, we perform a statistical analysis of QD surface measured by AFM (Fig. 3). The surface slopes *on QD surface* in two dimensions, i.e., $\partial h/\partial x$ and $\partial h/\partial y$) are collected and plotted in a form of two-dimensional histogram. The result for the GaSb QDs grown on $In_{0.25}Ga_{0.75}$ As is shown in Fig. 5(b). From this result, we see clearly the spot at the origin. This confirms a flat (001) surface of QD. Moreover, preferential facetted orientations of QD surface can also be extracted. They are $\{10n\}$ and $\{11n\}$, where n is a positive integer. Slightly misaligned of the indicated dotted lines for $\{10n\}$ and $\{11n\}$ is due to the slightly misalignment of the sample occurred during the AFM scan.

According to the statistical analysis shown above, we can draw a schematic of obtained GaSb/InGaAs QD shape as shown in Fig. 5(c). The QD consists of multi-facetted surface with a flat (001) on the top. This modelled QD shape might be important for further analysis of QD, i.e., for developing an overgrowth model [14] or a comparison with the results from other measurement techniques such as cross-sectional scanning tunnelling microscopy [12].

In order to utilize QDs in optoelectronic applications, QDs must be buried into a matrix. We therefore investigate the optical properties of capped GaSb QDs by using PL spectroscopy. We have interested in tuning the QD electronic properties by changing the indium content in InGaAs insertion layer. Both QD size and shape and PL peak energy are our measures. Consistency to our results from free-standing QD array is also observed from the PL measurement of capped samples. The capping process induces shape change and arsenic-intermixing into the capped QDs. PL reveals the optical characteristics of capped QDs. Figure 6 shows the PL spectra of capped GaSb/InGaAs QDs. It clearly shows the difference in the 20 K PL spectra under In-mole fraction of InGaAs insertion layer. PL of the capped samples shows the emission of QD peak at 1.2-1.3 eV. This corresponds to the emission from bulk $GaAs_{1-y}Sb_y$

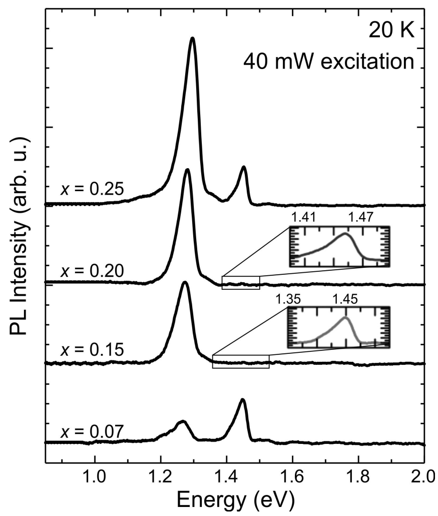

Fig.6: *PL measurement of capped GaSb QDs with 4-ML $In_xGa_{1-x}As$ insertion layer. Insets show the 10× magnification of the peaks around 1.45 eV.*

with the Sb content of only 11-17% [15]. However, this is an overestimated intermixing value because the quantum confinement and complicated band structure have not been taken into account. More realistic estimation of the Sb content in capped GaSb QDs needs more experimental evidence and/or sophisticated calculation techniques.

Concerning the line width of the QD peaks emitted at 1.2-1.3 eV, the line width shows *no* systematic change when the indium content of InGaAs insertion layer is varied. They are in the range of 55-63 meV. This non-systematic change and un-correlation with the height and diameter distribution of free-standing QDs (Fig. 4) might be due to the complication of the type-II band alignment and the intermixing process occurred during the capping. Overgrowth process is a subject for future experimental investigation.

According to the PL result (Fig. 6), the blueshift of the PL emission is observed when the indium content in InGaAs insertion layer increases. This is initially counter-intuitive because higher indium content should decrease the bandgap of the barrier layer. However, we can explain the result by the fact that the indium in the insertion layer does also alter the growth as well as the strain in the capped QD. The observed blueshift of PL emission from GaSb QDs is possibly due to the reduction of strain in the dots. This has been observed and reported without any clear explanation in Ref. [16]. This strain reduction results in the shift of the heavy-hole band and the quantized hole energy level [15]. Interestingly, relative PL intensity increases with the increasing indium content at the same excitation power (40 mW) and temperature (20 K). This indicates that the confinement of carriers in InGaAs insertion layer can possibly enhance the PL intensity, which relates to the carrier recombination in this nanostructure.

The unique characteristic of a type-II band alignment is seen when the laser power is varied [17]. As the density of photo-generated carriers in the dots increases, so does the strength of the Coulomb attraction to the electrons. Thus, as the excitation density is increased, the PL peak shifts to higher energy. This has been observed in GaSb/GaAs QDs. The expected behaviour is that the emission energy shows the third-root-of-excitation-intensity dependence [17, 18]. Figures 7(a) and 7(b) show comparative schematic of band diagram for type-II GaSb QDs when the carrier density is low (Fig. 7(a)) and high (Fig. 7(b)). Figure 7(c) shows an example of the normalized PL spectra of GaSb QDs grown on 4-ML $In_{0.25}Ga_{0.75}As$ insertion layer at 20 K with excitation intensities of 5 and 40 mW. The peaks at A and B are at 1.256 and 1.443 eV, respectively. The peaks at A' and B' are 1.28 and 1.448 eV, respectively. The peaks at A and A' are attributed to GaSb QD emission because of the exchange between antimony and arsenic on the dot surfaces during GaAs overgrowth or smaller

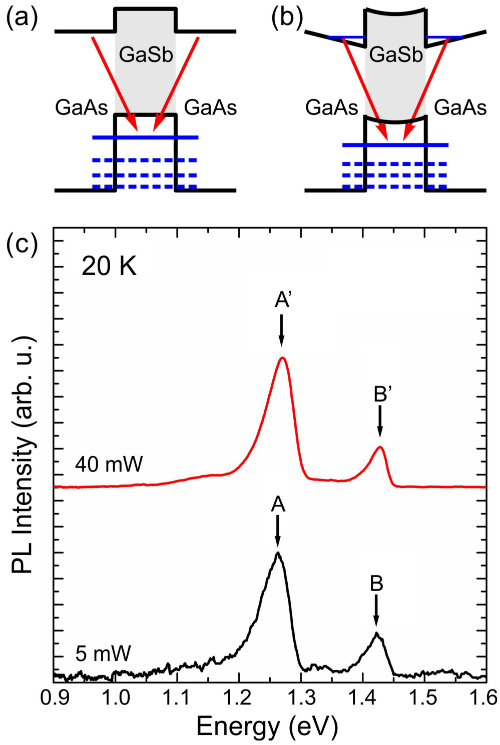

Fig. 7: *Schematics of the recombination in type-II QD nanostructure when carrier density is (a) low and (b) high. Band bending is due to Coulomb attraction between photo-generated electrons and holes. (c) The normalized PL spectra of GaSb QDs grown on 4-ML* $In_{0.25}Ga_{0.75}As$ *insertion layer at 20 K with an excitation intensity of 5 and 40 mW. Slightly blueshift of both peaks is observed.*

effective dot size after overgrowth [19, 20]. Slightly blueshift of the QD peak is observed when the excitation power increases. This observation is consistent with the theoretical characteristics of type-II QDs. We attribute the peaks B and B' at around 1.44 eV, which has been reported to occur in Si (As-site) with bulk GaAs, to other system-dependent impurities [21]. In this case (peak B-B'), the blueshift of the emission energy is also seen with increasing excitation intensity.

4. CONCLUSION

We have systematically investigated the effects of In-mole-fraction of InGaAs insertion layers on the structural and optical properties of the GaSb QDs grown on (001) GaAs substrate. Self-assembled growth process is used to realize GaSb QDs. The density of obtained GaSb QDs on InGaAs insertion layers is approximately $1.2\text{-}2.8\times10^{9}$ cm^{-2}. The uniformity of free-standing GaSb QDs is improved when the indium content increases. Observation of QDs reveals some preferential surface orientation of facetted QD with flat-top (001) surface. The PL peak energy and intensity can be altered by changing the indium composition of the InGaAs insertion layer. This indicates that the electron confinement in InGaAs insertion layer can modify the recombination in this structure. This work enhances the basic understanding of the structural and optical properties of GaSb QDs grown on InGaAs surface.

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Suwit Kiravittaya was born in Bangkok, Thailand, in 1976. He received the B.S. and Ph.D. degrees in Electrical Engineering from Chulalongkorn University in 1998 and 2003, respectively. After his Ph.D., he worked as a post-doc and became a scientist in Max-Planck Institute for Solid State Research, Stuttgart, Germany. In 2009, he became a scientist in the Institute for Integrative Nanoscience, IFW Dresden. Germany. Between May 2010 - October 2011, he led a research group named "Strained Nanomembranes for Optical, Electronic, and Fluidic Systems" in Institute for Integrative Nanoscience. Since 2012, he became a lecturer in the Department of Electrical and Computer Engineering, Faculty of Engineering, Naresuan University, Phitsanulok, Thailand. He is currently an assistant professor at Naresuan University. His current research interests are topics related to several advanced optical technologies including optical system for quantum information processing, novel opto-electronic devices. In 2015, he found a research unit so-called Advanced Optical Technology Laboratory.

Kamonchanok Khoklang was born in Phetchabun, Thailand, in 1990. She received the B.S. degrees in Advanced Materials and Nanotechnology Engineering from Silpakorn University in 2012 and M.S. degree in Electrical Engineering from Chulalongkorn University in 2015. From 2013 to 2014, she was with Osaka University as a research student by a supporting from JASSO, Japan. She current research interests and topics related to several advanced materials nanotechnology and optical technologies with semiconductor device.

Supachok Thainoi was born in Bangkok, Thailand, on October 12, 1956. He received the M.S. Degree in Nuclear Technology from Chulalongkorn University, Bangkok, Thailand in 1999. He is a Research Assistant in the Semiconductor Device Research Laboratory, Electrical Engineering Department, Faculty of Engineering, Chulalongkorn University. His current interests owe in the field of III-V Compound semiconductor and radiation detectors

Somsak Panyakeow received the B. Eng., M. Eng. and D. Eng. degrees, all in Electrical Engineering from Osaka University, Japan in 1969, 1971 and 1974 respectively. In 1974, he began to work at the Department of Electrical Engineering, Faculty of Engineering, Chulalongkorn University as a lecturer. He was a pioneer to set up the Semiconductor Device Research Laboratory (SDRL) at Chulalongkorn University in 1975. He was an Assistant Professor and an Associate Professor in 1977 and 1980 respectively. He was appointed to be a Full Professor of the department in 1982. He was promoted to be Professor C11 (the highest rank) in 1992. He has been engaged in research on photovoltaic devices and systems since 1975. He has been appointed to be Senior Research scholar by Thailand Research Fund (TRF) since 2005. In 2008, he was highly honored receiving prestigious "Dussadeemala" medal from his Majesty the King on his life-long research contribution. His recent research work is in the area of Molecular Beam Epitaxy for quantum devices and nanoelectronics & nanophotonics as well as for the development of high performance nanostructure solar cells.

Somchai Ratanathammaphan received the B.Eng, M.Eng and D.Eng. degree in Electrical Engineering from Chulalongkorn University, Thailand in 1984, 1988, and 1993, respectively. From 1986 to 1987, he was with Waseda University as a research student. From 1989 to 1990, he was with Tokyo Institute of Technology as a research student by a supporting from MEXT, Japan. During his doctoral work, he developed the combined technique of super-saturation and two-phase solution LPE growth of GaAs-GaAlAs single quantum well lasers by using a horizontal sliding boat with lateral sliding mechanism. In 1993, he joined the Department of Electrical Engineering, Chulalongkorn University as a lecturer. He became an assistant Professor and an Associate Professor in 2000 and 2003 respectively. Recently, he also has been engaged in the research of fabrication technologies of ultra-fine structure for nanoelectronics and photonics by molecular-beam epitaxy and the application of semiconductor nanowires on artificial photosynthesis and nanogenerators.