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# Molecular beam epitaxial growth of GaSb quantum dots on (0 0 1) GaAs substrate with InGaAs insertion layer



Kamonchanok Khoklang <sup>a</sup>, Suwit Kiravittaya <sup>b</sup>, Maetee Kunrugsa <sup>a</sup>, Patchareewan Prongjit <sup>a</sup>, Supachok Thainoi <sup>a</sup>, Somchai Ratanathammaphan <sup>a</sup>, Somsak Panyakeow <sup>a,\*</sup>

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#### ABSTRACT

We report on the molecular beam epitaxial growth of self-assembled GaSb quantum dots (QDs) on (0 0 1) GaAs substrates with an insertion layer. The insertion layer, which is a 4-monolayers (MLs)  $\ln_x Ga_{1-x} As$  ( $x=0.00,\ 0.07,\ 0.15,\ 0.20$  and 0.25), is grown prior to the QD growth. With this InGaAs insertion layer, the obtained QD density decreases substantially, while the QD height and diameter increase as compared with typical GaSb QDs grown on conventional (0 0 1) GaAs surface under the same growth condition. The GaSb QDs on GaAs have the dome shape with elliptical base and the elongation direction of the base is along the [1 1 0] direction. When the InGaAs insertion layer is introduced, the distinct elongation disappears and the QD sidewall shows facet-related surfaces with (0 0 1) plateau on top.

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## 1. Introduction

For a few decades, semiconductor quantum dots (QDs) have gained considerable interests due to their promising properties for device applications. Many electronic and optoelectronic devices based on III-V QDs have been investigated [1-6]. They include lasers, light emitting diodes, photodetectors, photovoltaic cells and memories. Among these devices, self-assembled In(Ga)As/GaAs QD is a major dot system that has been investigated. However, GaSb/ GaAs QDs, which have a type II band alignment, might be more suitable for some applications. For instance, GaSb/GaAs has been proposed to be used in charge-based memory device due to its large hole confinement [5]. Device performance will depend 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. In literature, different GaSb QD shapes have been reported [7,8]. For example, Jiang et al. [7] have shown that the elongation of GaSb QDs along the [1 1 0] direction can be controlled by changing the V/III ratio.

E-mail addresses: s\_panyakeow@yahoo.com, Somsak.P@chula.ac.th (S. Panyakeow).

http://dx.doi.org/10.1016/j.jcrysgro.2015.02.044 0022-0248/© 2015 Elsevier B.V. All rights reserved. In this work, we report on the structural properties of free-standing self-assembled GaSb QDs when  $\ln_x Ga_{1-x}As$  insertion layers with x=0.00, 0.07, 0.15, 0.20 and 0.25 are introduced. All samples were grown by a molecular beam epitaxy (MBE) with an Sb valved-cracker cell and investigated by atomic force microscopy (AFM). The presence of indium in  $\ln GaAs$  insertion layer induces the substantial decrement of QD density, the enlargement of QD size (both height and diameter) and the transition from elongated QD shape to QD with facet-related sidewalls and (0 0 1) plateau on top.

#### 2. Experiments

All samples were grown on semi-insulating (0 0 1) GaAs substrates in Riber Compact 21 solid-source MBE equipped with an Sb valved-cracker cell. After the deoxidation of surface oxide on GaAs substrate at 580 °C under As<sub>4</sub> atmosphere, GaAs buffer layer was grown at this temperature with the growth rate of 0.5 monolayer per second (ML/s). During the growth, reflection high-energy electron diffraction (RHEED) pattern was observed and well-prepared buffer layer ((0 0 1) GaAs surface) showed a clear (2 × 4) surface reconstruction. In order to grow the InGaAs insertion layer, the substrate temperature was ramped down to 500 °C. After the substrate temperature stabilized, 4-ML  $\ln_x$ Ga<sub>1-x</sub>As was grown. This thickness is chosen so as to have a flat InGaAs surface without any surface relaxation [9]. When growing this layer, indium and gallium

ล้ายนาถูกต้อง Grant Kirovittays

<sup>&</sup>lt;sup>a</sup> The Semiconductor Device Research Laboratory (SDRL), Department of Electrical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand

b Department of Electrical and Computer Engineering, Faculty of Engineering, Naresuan University, Phitsanulok 65000, Thailand

<sup>\*</sup>Correspondence to: The Semiconductor Device Research Laboratory (SDRL), Department of Electrical Engineering, Faculty of Engineering, Chulalongkorn University. Phayathai Road, Patumwan, Bangkok 10330, Thailand. Tel.: +66 2 218 6521; fax: +66 2 218 6523.

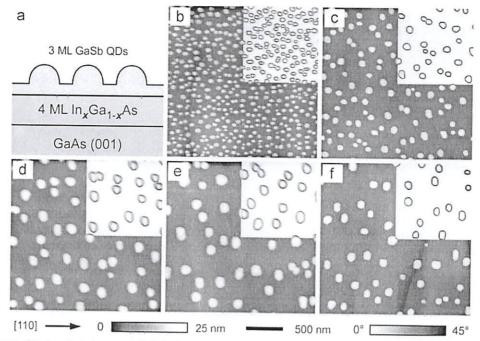


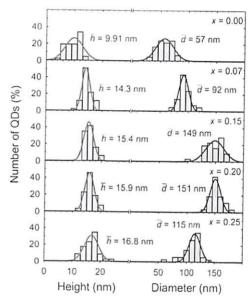
Fig. 1. (a) Schematic diagram of the investigated structure: GaSb QDs (3 ML) on top of 4-ML-thick  $In_xGa_{1-x}As$  insertion layer. (b)–(e)  $2 \times 2 \mu m^2$  AFM images of GaSb QDs on  $In_xGa_{1-x}As$  with x=0.00, 0.07, 0.15, 0.20 and 0.25, respectively. Insets show a portion of the same AFM images with surface slope scale.

growth rates are 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 rates are 0.025, 0.025, 0.025, and 0.033 ML/s and gallium rates are 0.33, 0.14, 0.10, and 0.10 ML/s, respectively. After the 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. After the growth, the sample was cooled down immediately. The surface morphology was characterized by an AFM (Seiko SPA-400) in dynamic force mode in air and the post-processing was done in MATLAB program.

# 3. Results and discussion

Fig. 1(a) shows a schematic diagram of investigated surface structure. Fig. 1(b)–(e) show  $2 \times 2 \ \mu m^2$  AFM images of GaSb QDs on GaAs (Fig. 1(b)) and on  $In_xGa_{1-x}As$  (Fig. 1(c)–(f)) surfaces. Insets of AFM images in Fig. 1(b)–(f) show a portion of the surface (obtained from the same AFM image) with surface slope scale [10]. Distinct surface morphology is observed when  $In_xGa_{1-x}As$  layer is introduced, i.e., QD density substantially decreases and QD size increases. Gradual morphology changes are still observed when the indium content increases from x=0.07 (Fig. 1(c)) to 0.25 (Fig. 1(f)). This is due to the presence of different amount of indium in the InGaAs insertion layer.

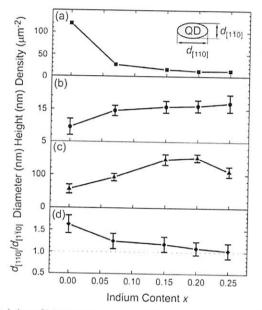
Analysis of height and diameter distributions of GaSb QDs on different InGaAs insertion layers is displayed as histograms in Fig. 2. The data are obtained from the analysis of *individual QD* in each AFM image (shown in Fig. 1(b)–(e)). First, the QD base area is extracted from the largest closed-loop contour line of contour plots of the AFM data. The QD diameter is then calculated with a circular base approximation. The QD height is obtained from the difference between the height level of the largest closed-loop contour line and the maximum height data. Histograms of QD height and QD diameter are then fitted with Gaussian functions. Fitted center positions together with the fitted curves are shown in Fig. 2. From this figure, one can clearly see that both QD height and diameter increase when the InGaAs insertion layer is



**Fig. 2.** Histograms of QD height and diameter distribution of GaSb QDs on  $\ln_x Ga_{1-x} As$  with x=0.00 (top), 0.07, 0.15, 0.20 and 0.25 (bottom). Solid lines are Gaussian function fits. Center position of the obtained Gaussian function fits are indicated in the figures.

introduced. Abrupt change is observed when the indium content is changed from x=0.0 (no insertion layer) to x=0.07. By further increasing the indium content, the QD height and 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. Indium adatoms might still present after 60-s Sb-soaking process because of their initially high content. Incorporating indium atoms into GaSb QDs induce high mismatch strain and enhance the growth of large aspect ratio QDs since the QD with a high ratio can relax more strain [11].

สาเนาถูกต้อง Gunt Kiravitlaya Fig. 3 summarizes the structural variation of GaSb QDs on the  $\ln_x Ga_{1-x}As$  insertion layer as a function of indium content in the insertion layer. When the  $\ln_x Ga_{1-x}As$  insertion layer is introduced, the QD density reduces, whereas QD height and diameter increase, as depicted in Fig. 3(a)–(c). The initial GaSb QD density, which is about  $120~\mu m^{-2}$ , decreases to less than  $30~\mu m^{-2}$ , while the average QD height increases about 1.5 times (from 9.5 nm for x=0.0 to 14.5 nm for x=0.07) when the  $\ln_{0.07}Ga_{0.93}As$  layer is inserted. These observations are attributed to the change of initial surface energy, interface energy as well as the strain energy of the QD system [11,12].



**Fig. 3.** Variations of (a) QD density. (b) average QD height. (c) average QD diameter and (d) degree of elongation (defined as  $d_{[1-1-0]}/d_{[1-1-0]}$ ) as a function of indium content x in  $\ln_x \text{Ga}_{1-x} \text{As}$  insertion layer. Inset of (a) shows the definition of  $d_{[1-1-0]}$  and  $d_{[1-1-0]}$ . The error bars in (b)–(d) are the standard deviation of the values.

To investigate the QD shape anisotropy, we define a degree of QD elongation as the ratio  $d_{[1\ 1\ 0]}/d_{[1\ \overline{1}\ 0]}$ , where  $d_{[1\ 1\ 0]}$  and  $d_{[1\ \overline{1}\ 0]}$  is the length of QD base along the [110] and [1 $\overline{1}\ 0$ ] direction, respectively. Inset of Fig. 3(a) shows a schematic diagram of a QD with these lengths. Fig. 3(d) displays the degree of elongation as a function of indium content obtained from the AFM data. The elongation of GaSb QD shape gradually changes from a clear elongation along the [110] direction to almost isotropic (no elongation) QD shape. This change can relate to the distinct  $\{1\ 1\ n\}$ -facet formation on QD surface when the InGaAs insertion layer is introduced and it will be explained below. It is noteworthy that this structural elongation influences the intrinsic properties of QDs such as their polarization dependency [7] and their transport properties [13].

For further quantifying the QD shape, a surface orientation mapping, which is so-called facet plot, is analyzed [10,14]. Fig. 4(a) shows the facet plot obtained from the QD surface of a GaSb QD without InGaAs insertion layer as compared that with In<sub>0.07</sub>Ga<sub>0.93</sub>As insertion layer (Fig. 4(b)). From these plots (and also the insets of Fig. 1(b)-(f)), we can clearly see that the (001) surface as a plateau on top of each GaSb QD is present when InGaAs insertion layer is introduced. This plateau is also observed in other AFM images of GaSb QDs grown on InGaAs surface (not shown). Apart from this plateau, other facet surfaces, which are likely to orient along the  $\{1 \ 1 \ n\}$  directions, are observed in the facet plot (see Fig. 4(b)). The facet-related surfaces are on the sidewall of the QDs. It is known from previous theoretical growth study that facet can form on the sidewall of QD due to its low surface energies [15]. This induces the so-called self-limiting growth, which stabilizes the QD size and shape. When indium content in the insertion layer increases, the sidewall facets tend to become steeper. However, due to the limited resolution of our AFM, we cannot fully identify the facet indices. Based on our investigation, we can draw simple illustrations for the shape of GaSb QD grown on GaAs and InGaAs surfaces as shown in Fig. 4(c) and (d), respectively. For GaSb QDs on GaAs, we obtain an elongated base QD with rather round shape, while the GaSb QDs on InGaAs show some facetted surfaces with the (001) plateau on top. Moreover, the height and diameter of GaSb QDs on InGaAs are larger than those on GaAs.

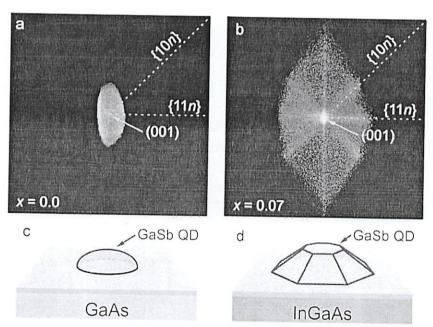


Fig. 4. Facet plots obtained from the QD surface of a GaSb QD (a) without and (b) with In<sub>0.07</sub>Ga<sub>0.93</sub>As insertion layer. (c) and (d) are illustrations of GaSb QD on (0 0 1) GaAs surface and on (0 0 1) InGaAs surface. Relatively small QDs, which are elongated along the [1 1 0] direction, are observed for GaSb growth on GaAs, while large facet-related CaSb QDs are formed when InGaAs insertion layer is introduced before the QD growth.

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#### 4. Conclusions

We present an investigation of structural morphology of self-assembled GaSb QDs on (0 0 1) GaAs substrates. We have shown that when the InGaAs insertion layer is introduced, the obtained QD morphology considerably changes. The QD density decrement as well as QD size (height and diameter) enlargement are observed. Besides, the degree of QD elongation changes from a distinct elongation along the [1 1 0] direction to an isotropic shape when the indium content in InGaAs insertion layer increases. Finally, we have shown that the GaSb QDs on InGaAs have a clear (0 0 1) plateau on top and facet-related surfaces on the sidewalls.

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#### References

- [1] V.M. Ustinov, A.E. Zhukov, N.A. Maleev, A.R. Kovsh, S.S. Mikhrin, B.V. Volovik, Y. G. Musikhin, Y.M. Shernyakov, M.V. Maximov, A.F. Tsatsul'nikov, N.N. Ledentsov, Zh.I. Alferov, J.A. Lott, D. Bimberg, 1.3 µm. InAs/GaAs quantum dot lasers and VCSELs grown by molecular beam epitaxy, J. Cryst. Growth 227-228 (2001) 1155–1161.
- [2] J. Sabarinathan, P. Bhattacharya, P-C. Yu, S. Krishna, An electrically injected InAs/GaAs quantum-dot photonic crystal microcavity light-emitting diode, Appl. Phys. Lett. 81 (2002) 3876–3878.

- [3] K. Hirakawa, S.-W. Lee, Ph. Lelong, S. Fujimoto, K. Hirotani, H. Sakaki, High-sensitivity modulation-doped quantum dot infrared photodetectors, Microelectron, Eng. 63 (2002) 185–192.
- [4] K. Janabe, D. Guimard, D. Bordel, Y. Arakawa, High-efficiency InAs/GaAs quantum dot solar cells by metalorganic chemical vapor deposition, Appl. Phys. Lett. 100 (2012) 193905.
- [5] M. Hayne, R.J. Young, E.P. Smakman, T. Nowozin, P. Hoidgson, J.K. Carleff, P. Rambabu, P.M. Koenraad, A. Marent, L. Bonato, A. Schliwa, D. Bimberg. The structural, electronic and optical properties of GaSb/GaAs nanostructures for charge-based memory, J. Phys. D: Appl. Phys. 46 (2013) 264001.
- [6] C.C. Tseng, W.-H. Lin, S.-Y. Wu, S.-H. Chen, S.-Y. Lin. The transition mechanisms of type-II GaSh/GaAs quantum-dot infrared light-emitting diodes, J. Cryst. Growth 323 (2011) 466–469.
- [7] C. Jiang, T. Kawazu, S. Kobayashi, H. Sakaki, Molecular beam epitaxial of very large anisotropic GaSb/GaAs quantum dots. J. Cryst. Growth 301-302 (2007) 828–832.
- [8] M. Kunrugsa, S. Kiravittaya, S. Sopitpan, S. Ratanathammaphan, S. Panyakeow, Molecular beam epitaxial growth of GaSb/GaAs quantum dots on Ge substrates, J. Cryst. Growth 401 (2014) 441–444.
- [9] G.L. Price, Critical-thickness and growth-mode transitions in highly strained In<sub>x</sub>Ga<sub>1-x</sub>As films, Phys. Rev. Lett. 66 (1991) 469-472.
- [10] S. Kiravittaya, A. Rastelli, O.G. Schmidt, Self-assembled InAs quantum dots on patterned GaAs(0.0.1) substrates: formation and shape evolution, Appl. Phys. Lett. 87 (2005) 243112.
- [11] V.A. Shchukin, D. Bimberg, Spontaneous ordering of nanostructures on crystal surfaces, Rev. Mod. Phys. 71 (1999) 1125–1171.
- [12] H. Eisele, M. Dähne, Critical thickness of the 2-dimensional to 3-dimensional transition in GaSb/GaAs(0.0.1) quantum dot growth, J. Cryst. Growth 338 (2012) 103–106.
- [13] G. Li, C. Jiang, H. Sakaki, Anisotropic scattering of elongated GaSb/GaAs quantum dots embedded near two-dimensional electron gas, J. Nanosci, Nanotechnol, 11 (2010) 10792–10795.
- [14] G. Costanini, A. Rasfelli, C. Manzano, R. Songmuang, O.G. Schmidt, K. Kern, K. von Känel, Universal shapes of self-organized semiconductor quantum dots: striking similarities between InAs/GaAs(0.0.1) and Ge/Si(0.0.1), Appl. Phys. Lett. 85 (2004) 5673–5675.
- [15] D.E. Jesson, G. Chen, K.M. Chen, S.J. Pennycook. Self-limiting growth of strained faceted islands. Phys. Rev. Lett. 80 (1998) 5156–5159.

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