

# MOVPE grown self-assembled InSb quantum dots: Structural characterization

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**Abstract.** InSb quantum dots were grown on GaSb and GaInSb surfaces using metal-organic vapour phase epitaxy (MOVPE) at atmospheric pressure. The effects of surface morphology and indium mole fraction in the buffer layer on the height of the islands were investigated. Quantum dots grown on GaSb (100) substrates etched for 120 s using a tartaric acid solution are flatter and have a lower surface density than those etched for 120 s using a hydrochloric acid solution. The indium mole fraction in the buffer layer was varied from 0 to ~0.08 in order to study the change in height of the dots. As the percentage of indium increases it leads to a reduction in lattice mismatch and thus lateral strain at the interface, resulting in a higher density of dots with smaller average height.

## 1. Introduction

In recent years there has been a tremendous development in the field of self assembled quantum dots (QDs) due to their unique physical and optical properties [1]. Self-assembled QDs are normally formed using the Stranski-Krastanov growth mode [2]. An example of a QD system, suitable for mid-infrared optoelectronic devices, is InSb in a GaSb matrix [3]. Most of the work performed to date on this system has been by molecular beam epitaxy (MBE) [4, 5]. Comparatively little work has been done on MOVPE grown structures, especially when considering InSb QDs grown on GaInSb surfaces. It has been shown theoretically that a change in the aspect ratio of InAs QDs grown on GaAs surface results from a change in the indium mole fraction in the InGaAs capping layer [6]. Since the lateral alignment, surface distribution, size and height of QDs play an important role in the design of mid-infrared detectors [7], it is important to test the influence of strain on these properties for the InSb/GaSb QD system.

In this paper the effects of the surface morphology of GaSb and GaInSb surfaces on the size and surface distribution of InSb QDs are reported. The effect of indium mole fraction in the buffer layer on the height of the QDs is also investigated.

## 2. Experimental procedure

The QDs were grown by MOVPE at atmospheric pressure, using a GaSb (100) substrate. Trimethylindium, triethylgallium and trimethylantimony were used as precursors. A 200 nm thick GaSb buffer layer was first grown at a constant susceptor temperature of 513<sup>0</sup>C (typical growth rate of ~1 nm/s). The temperature was then decreased to the QD growth temperature of 450<sup>0</sup>C and InSb was deposited at a V/III ratio of 7.9 for times of 5 s to 40 s. In order to study the effects of substrate morphology on the surface distribution and dot size, the samples were grown using the same procedures and parameters mentioned above, but the substrate was etched using different etchants as mentioned in table 1.

**Table 1.** Types of etchants and etch times used for etching GaSb (100) substrate

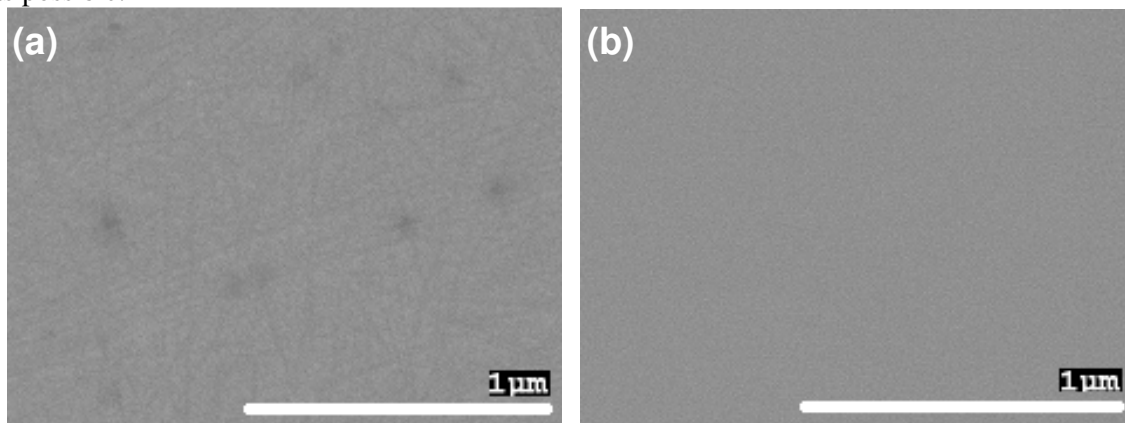
Etchant	Etch mixture	Etch time	Etch rate
Hydrochloric acid solution	HCl : H <sub>2</sub> O <sub>2</sub> (80 : 1)	120 s	~ 33 nm/s
Citric acid solution	aq. C <sub>6</sub> H <sub>8</sub> O <sub>7</sub> : H <sub>2</sub> O <sub>2</sub> (2 : 1)	120 s	~ 0.01 nm/s
Tartaric acid solution	H <sub>2</sub> C <sub>4</sub> H <sub>4</sub> O <sub>6</sub> : H <sub>2</sub> O <sub>2</sub> : HF (20 : 10 : 1)	120 s	~ 166 nm/s

In order to study the effects of indium mole fraction on the surface distribution and dot size, the samples were grown using the same procedures and parameters mentioned above, but with two different indium mole fractions of 0 and 0.3 in the vapour phase during growth of the 200 nm thick GaInSb buffer layer.

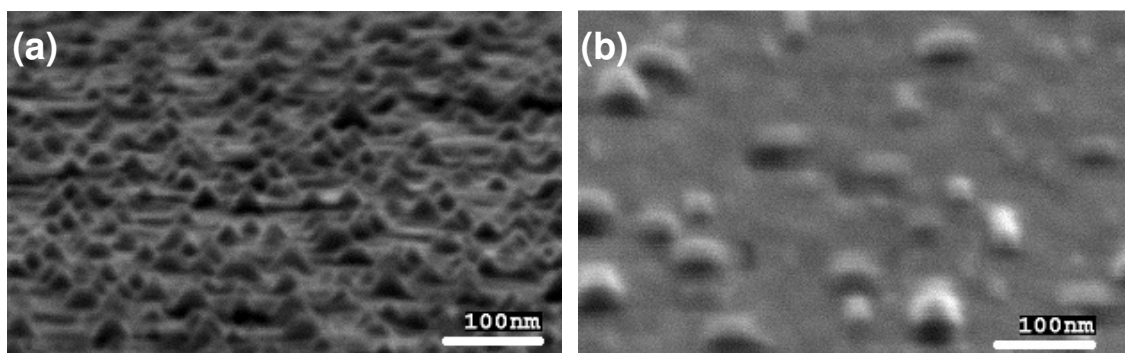
A JEOL 7001F field emission scanning electron microscope (SEM) was used to investigate the morphologies of the QDs. The elemental composition of the dots and buffer layer was analysed using a Perkin-Elmer ELAN 6100 inductively coupled plasma/mass spectrometer (ICP-MS). The surface features of the structures were studied using atomic force microscopy (AFM) using a SIS contact mode AFM housed on a CSM Instruments nano-indentor assembly. Strain and defects at the interface between the buffer and wetting layer were studied using a Philips CM20 transmission electron microscope (TEM).

### 3. Results and discussion

The surface morphology of the substrate plays an important role in surface migration and coalescence of quantum dots [8]. The influence of surface morphology on quantum dot height and surface density was studied by using different etchants to etch the GaSb (100) surface prior to growth. Three samples of GaSb substrate were etched for 120 s using the three solutions mentioned in Table 1. Figure 1 shows the surface morphology of the GaSb substrate surface etched with hydrochloric acid solution and tartaric acid solution for 120 s each. The tartaric acid solution gives the best surface morphology. The fine etch ( $\sim 4 \mu\text{m}$ ) obtained using the hydrochloric acid solution and superfine etch ( $\sim 1.2 \text{ nm}$ ) obtained using the citric acid solution is insufficient in smoothing out the roughness of the as-received substrate. It was found earlier that an etch depth of  $> 10 \mu\text{m}$  is required to achieve a clear improvement in the surface morphology [9]. It should be noted, however, that prolonged wet chemical etching has been found to produce a convex surface, hence the total etch depth needs to be kept as low as possible.

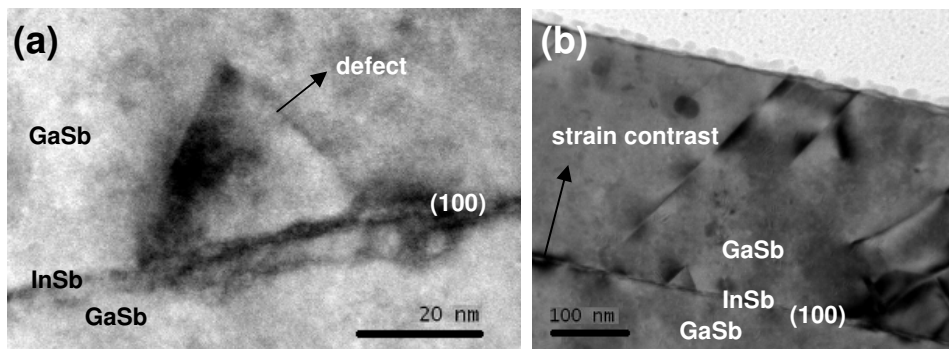


**Figure 1.** SEM images of GaSb (100) substrate surface etched in (a) hydrochloric acid solution and (b) tartaric acid solution for 120 s each.

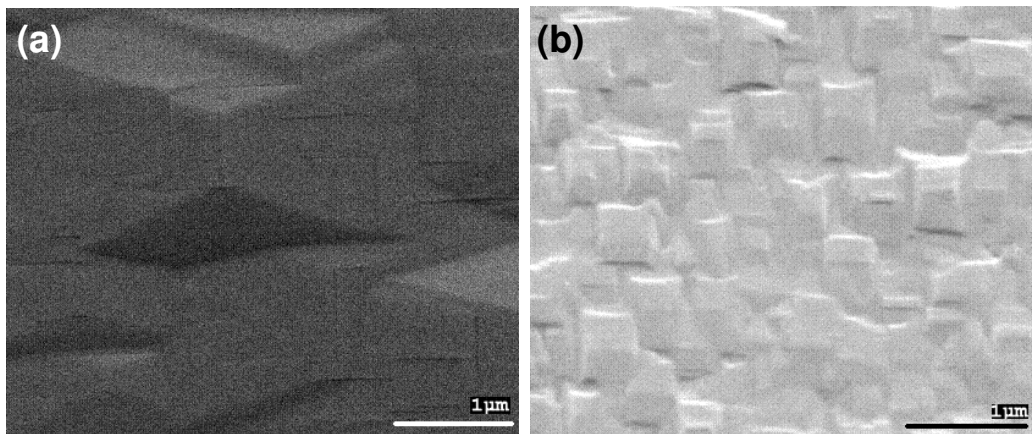


**Figure 2.** SEM images of InSb islands grown on GaSb (100) substrate surface etched in (a) hydrochloric acid solution and (b) tartaric acid solution for 120 s each.

In Figure 2 SEM micrographs are shown of samples in which InSb islands were grown for 6 s directly on an etched GaSb (100) surface using (a) the hydrochloric acid solution and (b) the tartaric acid solution. The density of dots observed for the substrate etched in the hydrochloric acid solution is considerably higher than for substrate etched in the tartaric acid solution. It is suggested that in the former case, the surface migration of indium atoms will be reduced compared to the latter case (due to the high density of nucleation sites), leading to a high density of small sized islands. The improved morphology of the surface after etching in the tartaric acid solution will promote surface diffusion; reduce the nucleation density, leading to a lower density of dots, as seen in Figure 2(b). The shape of the islands is also different: the pyramids shown in Figure 2(b) are truncated (“flatter”). The defects and strains at the interface were also investigated. In Figure 3(a) and 3(b) cross-sectional TEM micrographs of the interface between the InSb layer and GaSb (100) substrate are shown. The lattice mismatch of 6.3 % between the InSb wetting layer and the GaSb buffer layer can be clearly seen by the strain contrast images near the interface. Planar defects in the form of stacking faults can also be seen in Figure 3(a). The stacking faults are due to atomic stacking errors occurring during growth as a result of the lattice misfit [4].



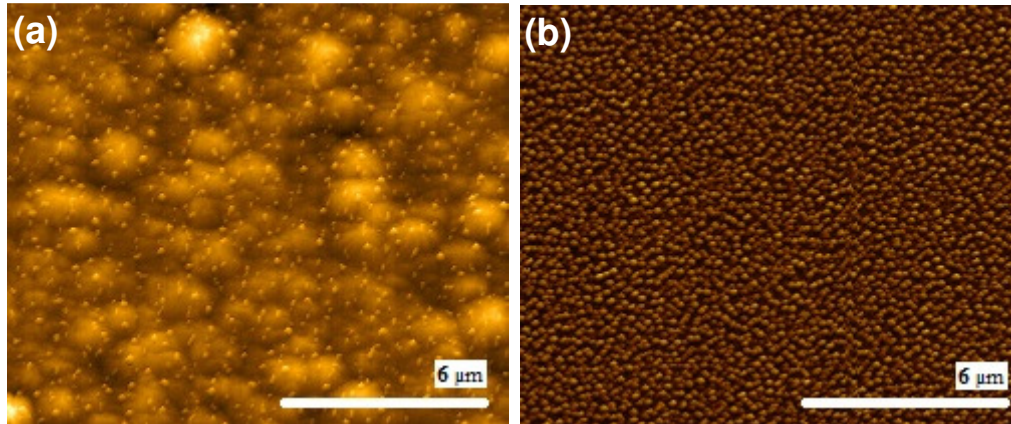
**Figure 3.** TEM images of InSb in a GaSb matrix showing (a) planar defects (b) strain contrasts at interface.



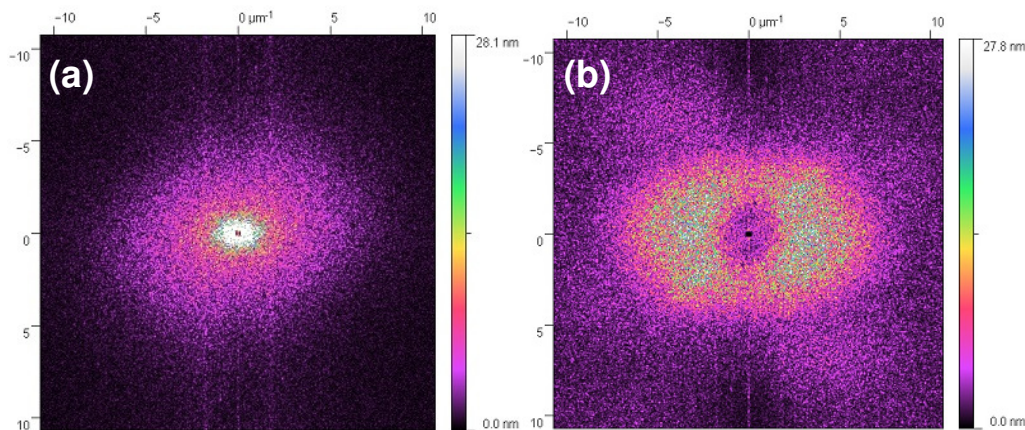
**Figure 4.** SEM images of surface of (a) ~ 200 nm GaSb and (b) ~ 200 nm GaInSb grown on GaSb (100).

In Figure 4(a) and (b) SEM micrographs of the surfaces of ~200 nm thick GaSb and GaInSb buffers can be seen. The buffer layers were grown under nominally identical growth conditions on substrate etched in the tartaric acid solution, but the surface morphology of the GaSb buffer is better than that of the GaInSb buffer due to an exact lattice match in the former case. In both cases the surface is uneven; the hills present on the GaSb surface, however, are larger in size as compared to those on the GaInSb surface. The facets of these broader hills act as flat surfaces for the nucleation and growth of InSb islands of size < 50 nm, which will lead to a lower density of dots. This is evident from the AFM images and accompanying fast Fourier transforms (FFT) of the AFM images shown in Figure 5 and 6. The intensity shown on the FFTs is a representation of the density and spatial separation between

dots. It is seen from the FFTs that the density of islands grown on GaSb is lower than those grown on GaInSb. Due to less strain between InSb/GaInSb as compared to between InSb/GaSb, the grown islands also have a lower height. Statistical analysis performed on the AFM images revealed an average QD height of 29 nm and 54 nm for samples grown on GaInSb and GaSb, respectively. An approximation of the indium content in the GaInSb buffer layer was done using ICP-MS chemical analysis, yielding a value of  $\sim 8\%$  (in the solid phase).



**Figure 5.** AFM images of InSb islands grown on (a)  $\sim 200$  nm GaSb and (b)  $\sim 200$  nm GaInSb buffer, both grown on GaSb (100).



**Figure 6.** FFT AFM images depicting surface density of InSb islands grown on (a)  $\sim 200$  nm GaSb and (b)  $\sim 200$  nm GaInSb buffer, both grown on GaSb (100).

#### 4. Conclusions

Single layer InSb self-assembled QDs were grown at  $450^{\circ}\text{C}$  using atmospheric pressure MOVPE on GaSb and GaInSb surfaces. The effects of substrate etching and indium mole fraction in the underlying buffer layer on the areal density of the QDs were investigated. QDs have a higher density on a substrate etched with hydrochloric acid solution, due to the high density of nucleation sites on such a relatively rough surface. As the indium concentration in the buffer layer increased, the lateral strain between the buffer and the dots was reduced, leading to a reduced height of the islands. This is a successful step towards the growth of multilayer QDs with strain engineering using spacer layers for detector applications.

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### References

- [1] V.A. Shchukin, N.N. Ledentsov and D. Bimberg 2004 *Epitaxy of Nanostructures*, Springer, Berlin.
- [2] D. Bimberg, M. Grundmann and N. Ledentsov 1999 *Quantum Dot Heterostructures*, John Wiley and Sons, England.
- [3] A. Krier 2006 *Mid-infrared Semiconductor Optoelectronics*, Springer, Berlin.
- [4] N. Deguffroy , V. Tasco, A.N. Baranov, E. Tournie, B. Satpati, A. Trampert, M.S. Dunaevskii, A. Titkov and M. Ramonda 2007 *J. Appl. Phys.* **101** 124309.
- [5] V. Tasco, N. Deguffroy, A.N. Baranov, E. Tournie , B. Satpati, A. Trampert, M. Dunaevski and A. Tiktov 2006 *Appl. Phys. Lett.* **89** 263118.
- [6] M. Usman, D. Vasileska and G. Klimeck 2010 *Physics of Semiconductors* **1119** 527.
- [7] R. Paiella 2006 *Intersubband Transitions in Quantum Structures*, McGraw Hill, USA.
- [8] P. Sutter, E. Mateeva-Sutter and L. Vescan 2001 *Appl. Phys. Lett.* **78** 1736.
- [9] M. Godbole, E.J. Olivier, E. Coetsee, H.C. Swart, J.H. Neethling and J.R. Botha 2011 submitted to *Physica B: Condensed Matter* (under review).