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Fundamental Study of Antimonide Nanostructures by Molecular Beam Epitaxy

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The research objective was to conduct experimental work in molecular beam epitaxial growth of GaSb/GaAs and InSb/GaAs quantum dots (QDs) are conducted and compared with conventional InAs/GaAs QDs on (001) Ge substrates and on (001) GaAs substrates for comparison.

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Executive Report

on

Fundamental Study on Antimonide Nanostructures by Molecular Beam Epitaxy
Supported by AOARD 2014
(Award No.FA2386-14-1-4081)

by

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Abstract

This is a report on research activities during 11 months of the project supported by AOARD from September 2014 to July 2015 being conducted at Chulalongkorn University in Thailand. Following the research work on InAs quantum dots (QDs) and quantum dot molecules (QDMs) grown by molecular beam epitaxy (MBE), the research target is extended to GaSb QDs and InSb QDs which are type II quantum nanostructures. This type II nanostructure would have a unique property of separated carrier confinement for novel nanoelectronic devices such as IR detectors, solar cells, memory devices, etc.

Prior to AOARD support in 2014, new MBE machine (RIBER compact 21) with Antimony (Sb) cracker cell has been installed by financial supports from Thailand Research Fund (TRF), Nanotechnology Center of Thailand and Chulalongkorn University.

GaSb QDs are intensively investigated for their formation mechanism on GaAs and Ge substrates. Photoluminescence measurement is also conducted to study on their optical property reflecting their dot size, dot density and dot uniformity which are basic information for future device design.

Droplet epitaxy technique is also developed for GaSb quantum rings (QRs) formation which will be useful for spintronic devices.

InSb QD is another quantum nanostructure being studied. InSb is the narrowest bandgap semiconductor and is useful for long infrared wavelength detection. InSb QDs on InAs is type III nanostructure of which interesting electron transport could be foreseen for nanoscale memory devices. This part of preliminary research activity was not proposed in 2014 but will be added to the research proposal in 2015.

Introduction

Mid- and far-infrared wavelengths are useful for night vision and gas pollution detection. Most of infrared detectors work at low temperature. Sb based QDs are candidates for infrared detection. Due to their quantum nanostructure, they could be operated at high temperature and are more stable with temperature change.

Sb based QDs can be type II and type III nanostructures which would be applied for memory devices due to their long carrier life time [1-3].

GaSb/GaAs and InSb/GaAs have 7.8% and 14.6% lattice mismatch providing some growth conditions to form quantum dots by S-K growth mode similar to InAs/GaAs QDs having 7.2% lattice mismatch.

There are several research motivations in this work. Ge is a well known bulk material for infrared detectors. Au doped Ge, Hg doped Ge, Cu doped Ge are used for mid- and far-infrared applications. They are useful detectors for CO_2 lasers. However, they must be operated at low temperature using liquid Nitrogen or liquid Helium.

Sb based QDs are also candidates for infrared detection. Their nanostructures as well as their band gaps can be controlled for specific wavelength sensitivity. However, GaSb/GaAs and InSb/GaAs QDs are type II and InSb/InAs QD is type III comparing to InAs/GaAs QD which is type I. Therefore, GaSb/GaAs and InSb/GaAs QDs and InSb/InAs QDs can give interesting behavior in their electro-optic properties due to separated confinements of electrons and holes in type II and type III nanostructures [4].

Ge and GaAs is mostly lattice match. Good GaAs epitaxial layer can be grown on Ge substrate with 6° miscut. Ge is non-polar material. When a polar material like GaAs is epitaxially grown on (001) Ge substrate, Anti-Phase Domains (APDs) are created [5]. Domain boundary is a kind of defects which might affect the optical and electrical properties of the semiconductor material. However, when the GaAs domain size is large comparing the nanostructure of Sb based QDs, their unique properties should remain and are useful for nano-photonic device applications. Therefore, (001) Ge substrates can be used in our research work.

Combination of Ge and Sb based QDs for infrared absorption is the major motivation of this research work.

References

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[2] K. Suzuki, R.A. Hogg and Y. Arakawa, *Journal of Applied Physics* **85**, 8349 (1999).

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[4] A. Cho, MBE Pioneer's Invited Talk at *ICMBE-2014*, Flagstaff, Arizona, 6-12 September (2014)

[5] M. Kunrugsa, S. Kiravittaya S. Sopitpan, S. Ratanathammaphan, and S. Panyakeow, *Journal of Crystal Growth*, Vol. **401**, pp. 441-444 (2014)

Research Outputs

Sample Preparation

In our experimental work, molecular beam epitaxial growth of GaSb/GaAs and InSb/GaAs quantum dots (QDs) are conducted and compared with conventional InAs/GaAs QDs on (001) Ge substrates and on (001) GaAs substrates for comparison. The schematic structure of the samples is shown in figure 1.

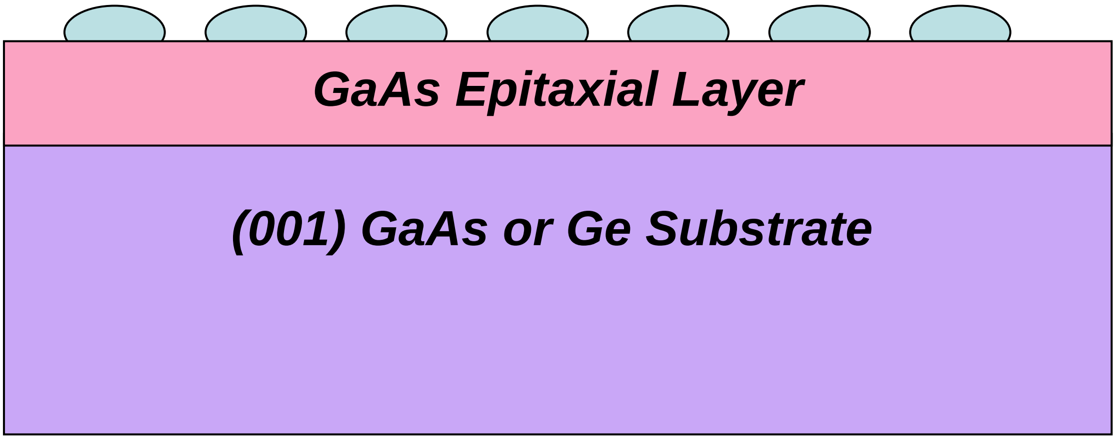


Figure 1 Sb based QDs on GaAs epitaxial layer grown on (001) GaAs or Ge substrate

InAs/GaAs QDs growth process on (001) GaAs or Ge substrate is conducted with standard growth condition. The experimental work is conducted by conventional growth of GaAs buffer layer on (001) GaAs or Ge substrate at 580°C with growth rate of 0.5 ML/s.

Then, the substrate temperature is reduced to 500°C for InAs QDs formation by S-K growth technique. InAs QDs are grown with the growth rate of 0.01 ML/s. As_4 flux is kept at $8x10^{-6}$ Torr throughout the growth process.

GaSb/GaAs and InSb/GaAs QDs are grown on (001) Ge substrates and on (001) GaAs substrates for comparison by the following steps. The growth process starts from an epitaxial growth of GaAs buffer layer with thickness of 500 nm at 580°C. Then, Ga shutter is shut off and the substrate temperature is lowered to either 450°C for GaSb QD formation or 300°C for InSb QD formation.

When the temperature becomes stable at either 450°C or 300°C , As shutter is closed until the background pressure reaches 10^{-9} Torr. Sb cracker cell working at 700°C for stable flux pressure $>6x10^{-7}$ Torr is opened with the soaking time of 1 minute, then either Ga shutter (22 sec) or In shutter (>60 sec) is opened again for GaSb or InSb QDs formation. Spotty RHEED patterns are confirmed when QDs are formed.

Experimental Result:
AFM Images of InAs, GaSb and InSb QDs on (001) Ge Substrates

Figure 2 shows the AFM images of InAs, GaSb and InSb QDs on (001) Ge substrates.

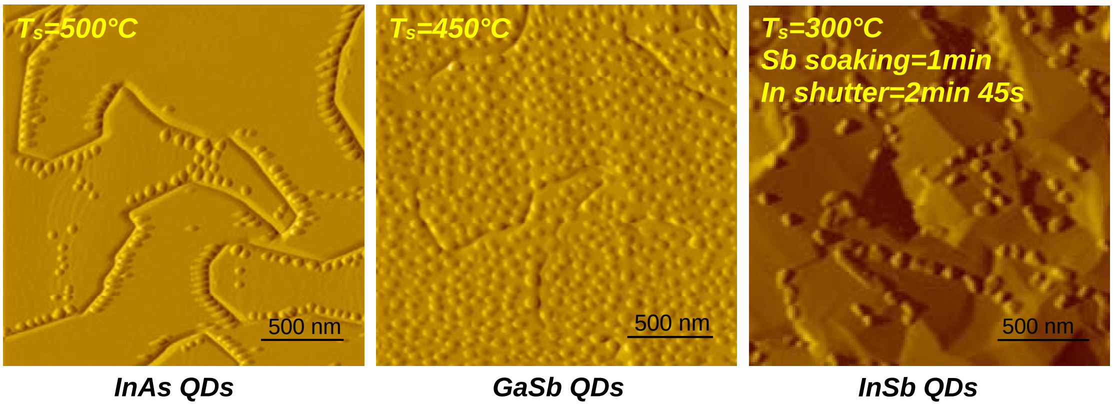


Figure 2 AFM Images of InAs, GaSb and InSb QDs on (001) Ge Substrates

Anti-phase domains can be observed on all samples. InAs and InSb QDs are found at APD boundary while GaSb QDs are scattered on the whole surface of APDs.

Comparison between InAs QDs and InSb QDs on GaAs APDs/Ge substrates

InAs QDs are formed at edge of domain boundary. They are elongated along [1-10] which is perpendicular to the line of boundary. In the contary, InSb QDs are formed right on the domain boundary. They are elongated along [110] which are parallel to the line of boundary as shown in figure 3.

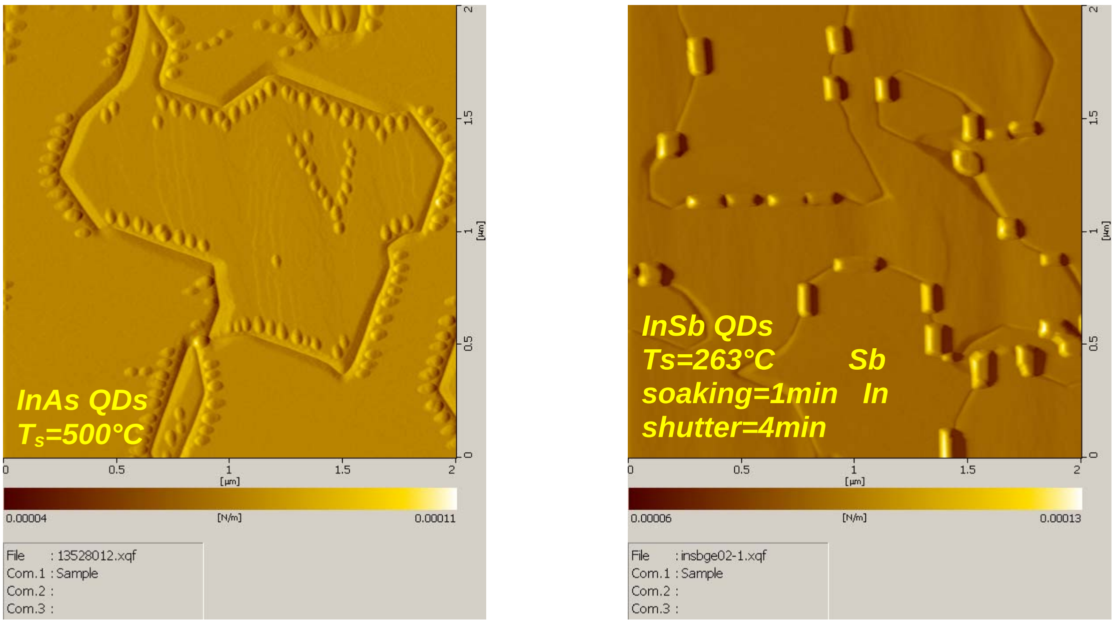
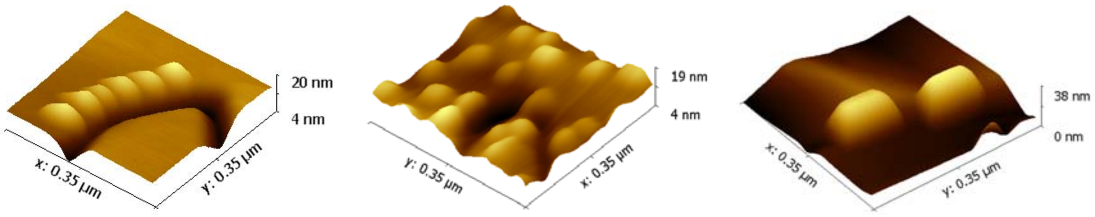


Figure 3 Comparison between InAs QDs and InSb QDs on GaAs APDs/Ge substrates

Dot Morphologies grown on GaAs APDs/Ge



InAs QD ***GaSb QD*** ***InSb QD***

Figure 4 Dot morphologies of InAs, GaSb and InSb QDs on GaAs APDs/Ge

InAs dots have an elongated oval shape along [1-10] crystallographic direction and are situated only on one side of each APD grain. In contrast, GaSb dots have more symmetrical dome shape and are scattered on the whole surface of the sample. New finding is the peculiar shape of InSb dots. InSb dots have a rectangular shape with sharp facets and they are situated along the line [110] of APD grain boundary.

Discussion of the Results:

Discussion on Growth Mechanism

Sb is less mobile atom, therefore, GaSb/GaAs QDs are distributed on the whole surface of GaAs epitaxial layer regardless the existence of APD boundary. GaSb dot density per unit area is high (> 10×10 cm^{-2}) which is useful for nano-photonic device applications.

On the other hand, it is found that InSb/GaAs QDs behave differently. The InSb QD formation is similar to that of InAs/GaAs QDs grown on GaAs/Ge epitaxial layer, where As-rich atmosphere is applied. Arsenic is a fast diffused atom. This is how InAs QDs are preferable to be created at the edge of APD boundary or on the line of boundary where strain is accumulated. The similarity between InAs and InSb QD formation indicates that In atoms are more influential species than Sb atoms that control the dot formation mechanism.

It is found in our MBE growth process that InSb QDs are not created at high temperature range of 400-500°C. Therefore, growth temperature of InSb QDs is set up at 300°C. However, dot formation needs long In deposition time of more than 130 seconds due to some relaxation prior to the creation of InSb dots at the line of domain boundary.

When Indium is incorporated in both Sb and As based QDs, dot formation mechanisms are similar, but not in the case of Gallium.

Due to high sticking coefficient of Sb atoms deposited during Sb soaking time, GaSb QDs are formed on the whole surface of GaAs APDs indicating that Ga atoms are less mobile than In. atoms.

Discussion on Type of Quantum Dots

Due to different band gaps and different electron affinities of dot materials as well as their lattice mismatch strain related to those of GaAs, different types of quantum dots are formulated.

InAs/GaAs dot is type I with both electrons and holes are confined at the quantized states of the dot. GaSb/GaAs and InSb/GaAs dots are type II with only holes in the quantized state but electrons are separated in the bulk GaAs. InSb/InAs dot is type III having similar separated confinement to type II but with broken gap, therefore, tunneling effect is possible in the carrier transport.

When electrons are separated in bulk GaAs from holes in quantized levels of Sb based QDs. This leads to long carrier life time of type II GaSb dots and type II InSb dots. This unique behavior is required for memory applications. Long carrier lifetime gives lower recombination rate of photo-excited carriers. This is another requirement for solar cell applications.

Summary

1. InAs/GaAs, GaSb/GaAs and InSb/GaAs quantum dots are experimentally grown on (100) Ge substrates and (001) GaAs substrates for comparison at respective MBE growth conditions.
2. Dot creation mechanism as well as their sites on GaAs APD/Ge and morphologies are compared and discussed.
3. Different types of quantum dots (Type I, Type II and Type III) are discussed.
4. Type II and Type III dot nanostructures have unique potential applications for nano-electronic, nano-photonic and memory devices due to separated carrier confinement and long carrier life time.

1. List of International Conference Papers (7 papers)

1.1) "GaSb/GaAs, InSb/GaAs & InAs/GaAs Quantum Dots on Ge for Nanophotonic Devices", Somsak Panyakeow, Suwit Kiravittaya, Supachok Thainoi, Songphol Kanjanachuchai, and Somchai Ratanathammaphan, **32nd International Conference on Physics of Semiconductors (ICPS-2014)**, Austin, USA., 10-15 August, 2014.

1.2) "Lateral Quantum-Dot Molecules Growth by Droplet Epitaxy", Nattapa Prapasawad, Somchai Ratanathammaphan, Patchareewan Prongjit, and Somsak Panyakeow, **32nd International Conference on The Physics of Semiconductors (ICPS-2014)**, Austin, USA., 10-15 August, 2014.

1.3) "Fabrication of GaSb Quantum Rings on GaAs (001) by Droplet Epitaxy", Maetee Kunrugsa, Kar Hoo Patrick Tung, Aaron James Danner, Somsak Panyakeow, and Somchai Ratanathammaphan, **18th International Conference on Molecular Beam Epitaxy (ICMBE-2014)**, Flagstaff Arizona, USA., 6-12 September, 2014.

1.4) "Molecular Beam Epitaxial Growth of GaSb Quantum Dots on (001) GaAs Substrates with InGaAs Insertion Layers", Kamolchanok Khoklang, Suwit Kiravittaya, Supachok Thainoi, Somchai Ratanathammaphan, and Somsak Panyakeow, **18th International Conference on Molecular Beam Epitaxy (ICMBE-2014)**, Flagstaff, Arizona, USA., 6-12 September, 2014.

1.5) "Gallium Antimonide Ring-With-Dot Structures Grown by Droplet Epitaxy", Maetee Kunrugsa, Somsak Panyakeow, Somchai Ratana Thammaphan, **18th European Molecular Beam Epitaxy Conference (EUMBE-2015)**, Canazei, Italy, 15-18 March, 2015.

1.6) "Effect of In-Amount in InAs Quantum-Dot Growth Step on Lateral InAs Quantum-Dot-Molecules Grown by 2-Step Growth Technique Using

Molecular Beam Epitaxy", Nattapa Prapasawad, Patchareewan Prongjit, Somsak Panyakeow, Somchai Ratanathammaphan, **ICMAT2015 & IUMRS-ICA2015**, Singapore, 28 June-3 July, 2015.

1.7) "Effect of InGaAs Insertion Layers on the Structural and Optical Properties of GaSb Quantum Dots", Kamonchanok Khoklang, Suwit Kiravittaya, Supachok Thainoi, Somsak Panyakeow, Somchai Ratanathammaphan, **ICMAT2015 & IUMRS-ICU2015**, Singapore, 28 June, 2015.

2. List of International Journal Publications (2 papers)

2.1) "Fabrication of GaSb Quantum Rings on GaAs (001) by Droplet Epitaxy", Maetee Kunrugsa, Kar Hoo Patrick Tung, Aaron James Danner, Somsak Panyakeow, and Somchai Ratanathammaphan, **Journal of Crystal Growth**, Vol. 425, pp. 287-290 , 2015.

2.2) "Molecular Beam Epitaxial Growth of GaSb Quantum Dots on (001) GaAs Substrates with InGaAs Insertion Layers", Kamolchanok Khoklang, Suwit Kiravittaya, Maetee Kunrugsa, Patchareewan Prongjit, Supachok Thainoi, Somchai Ratanathammaphan, and Somsak Panyakeow, **Journal of Crystal Growth**, Vol. 425 , pp. 291-294 , 2015.

3. List of Invited Talks and Keynote Lectures (5)

3.1) "Perspective of Research on Solar Cell and Nanoelectronics", **(Invited Talk)**, Somsak Panyakaow, International Conference on Safe and Sustainable Nanotechnology (in conjunction with 4th German-Thai Symposium on Nanoscience and Nanotechnology), Akatosarot Building, Naresuan University, Phitsanulok, Thailand, 14-17 October, 2014.

3.2) "Nanoelectronics and Nanophotonics-1", **(Invited Talk)**, Somsak Panyakeow, Nano-engineering Program, International School of Engineering (ISE), Faculty of Engineering, Chulalongkorn University, Bangkok, Thailand, 7 November, 2014.

3.3) "Nanoelectronics and Nanophotonics-2", **(Invited Talk)**, Somsak Panyakeow, Nano-engineering Program, International School of Engineering (ISE), Faculty of Engineering, Chulalongkorn University, Bangkok, Thailand, 14 November, 2014.

3.4) "Solar Cells and Their Potential Applications in Thailand", **(Invited Talk)** Somsak Panyakeow, Rajamongkol University (Suwannabhumi Campus), Ayudhaya, Thailand, 28 November, 2014.

3.5) "Global Perspective on Nanoelectronics and Their Future Applications", **(Keynote)**, Somsak Panyakaow, The Fifth International Conference on Science and Engineering (ICSE 2014), Inya Lake Hotel, Yangon, Myanmar, 30 December, 2014.