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Zhiming M. Wang
Editor

Self-Assembled Quantum Dots

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Self-Assembled Quantum Dots

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Chapter 12

In(Ga)As/GaAs Quantum Dots Grown by MOCVD for Opto-electronic Device Applications

K. Sears, S. Mokkalapati, H. H. Tan, and C. Jagadish

12.1 Introduction

This chapter focuses on the self-assembled growth of In(Ga)As/GaAs quantum dots using Metal Organic Chemical Vapor Deposition (MOCVD) and their application to quantum dot lasers and photonic integrated circuits.

Quantum dots (QDs) have attracted intense interest over the past decade due to their 3D confinement of carriers and unique properties. One application that has gained much attention is the semiconductor QD laser. Quantum dots offer a number of potential benefits for laser performance such as improved efficiencies, improved thermal stability and reduced threshold currents (Arakawa and Sakaki 1982, Dingle and Henry 1976). Since the first demonstration of a quantum dot laser in 1994 (Kirstaedter et al. 1994), many of these predicted advantages have been realized. For example, low threshold currents on the order of 20 A/cm² per dot layer (Liu et al. 2000, Sellers et al. 2004, Kovsh et al. 2002) and high thermal stability with characteristic temperatures as high as 300 K at room temperature (Kovsh et al. 2002, Shchekin et al. 2002, Maximov et al. 1997) have been reported. Lasing at 1.3 and 1.5 μ m has also been achieved by capping InAs quantum dots with thin InGaAs capping layers (Sellers et al. 2004, Karachinsky et al. 2005, Fehse et al. 2003, Ledentsov et al. 2003, Zhukov et al. 1999b). However it is still difficult to achieve all of the above characteristics in the one device, with only a few such reports (Kovsh et al. 2002, Shchekin et al. 2002). Furthermore, the majority of QD laser studies have focused on growth using molecular beam epitaxy (MBE). In comparison, there are very few reports of In(Ga)As/GaAs quantum dot lasers grown using metal-organic chemical vapor deposition (MOCVD) (Maximov et al. 1997, Sellin et al. 2001, Kaiander et al. 2004, Sellin et al. 2003, Heinrichsdorff et al. 1997b, Tatebayashi et al. 2005, Heinrichsdorff et al. 2000, Tatebayashi et al. 2003, Nuntawong et al. 2005, Walter et al. 2002, Sears et al. 2007, Lever et al. 2004a) and the production of high performance, long wavelength quantum dot lasers using MOCVD remains an elusive goal. This is despite the fact that MOCVD is the growth technique of choice for light emitting devices because of its scalability to high volume production and its ability to grow a diverse range of semiconductor compounds.

This chapter starts with a brief description of the MOCVD growth technique. The Stranski-Krastanow growth mode used for the self-assembled growth of In(Ga)As QDs is then described followed by a detailed discussion of the key MOCVD growth parameters and their influence on the nucleation process. Stacked QD growth is necessary for many device applications and this is reviewed in Section 12.4 before discussing the important lasing characteristics of QD lasers.

The integration of QD lasers with other components such as waveguides and modulators is also of interest. Such photonic integrated circuits (PICs) offer a number of advantages such as:

- reduced volume,
- reduced packaging costs,
- improved reliability, and
- reduced losses associated with fiber coupling.

A promising technique for the production of QD photonic integrated circuits is selective area epitaxy (SAE). Unlike other post growth processing techniques, SAE does not alter the quality of the QD active region which is a distinct advantage for active devices such as QD lasers. The final sections of this chapter discuss the basic principles of SAE and demonstrate how it can be used to achieve controlled nucleation and band gap tuning of QDs.

12.2 Basic Principles of Metal Organic Chemical Vapor Deposition (MOCVD)

There are two main epitaxial growth techniques used to grow self-assembled III-V semiconductor quantum dots. These are Metal Organic Chemical Vapor Deposition (MOCVD), also known as metal organic vapor phase epitaxy (MOVPE), and Molecular Beam Epitaxy (MBE). These techniques differ considerably in the sources used and surface growth kinetics. MOCVD is particularly complex with many factors such as precursor chemistry, growth kinetics, hydrodynamics, mass transport and surface chemistry influencing growth.

All of the samples and devices discussed in this chapter were grown on semi-insulating (001) GaAs substrates using a low pressure (100mbar) AIXTRON 200/4 horizontal flow MOCVD system. The MOCVD growth technique uses the group III metal alkyls (e.g. trimethyl-gallium, $\text{Ga}(\text{CH}_3)_3$) and group V hydrides (e.g. arsine, AsH_3) to grow thin layers of III-V semiconductor materials. These precursors flow over a heated substrate where they react to form a solid. As an example, the overall reaction for GaAs can be written as:

By mixing different metal alkyls and hydrides a wide range of binary, ternary and even quaternary III-V semiconductors can be formed. MOCVD is therefore highly

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versatile and capable of growing a wide range of III-V semiconductor compounds including both P and Al containing ones. The growth rate is generally dictated by the group III flow rate.

Figure 12.1 shows a schematic of the system used and the available sources. The sources used to grow the samples discussed in this chapter are trimethyl-indium (TMIn), trimethyl-gallium (TMGa), trimethyl-aluminium (TMAI) and arsine (AsH_3). Silane (SiH_4) and carbon tetrachloride (CCl_4) were used for n and p type doping, respectively. Diethyl-zinc (DEZn) is also often used as a p-type dopant. However Zn diffuses rapidly in GaAs and therefore CCl_4 was used instead.

The metal-organics (with the exception of trimethyl-indium) are liquids and are kept in bubblers. Palladium purified hydrogen is used as a carrier gas and is bubbled through the sources. The amount of metal-organics 'picked up' by the carrier gas is determined by the bath temperature and source pressure. The hydrides are stored in high pressure cylinders and supplied directly to the reactor. Mass flow controllers are used to deliver accurate gas flows to the reaction chamber. In order to avoid precursor pre-reactions, the hydrides and metal-organics are delivered separately in the Hydride and MO run lines, respectively, before entering the reaction chamber. In the reaction chamber, the substrate sits on a gas foil rotation plate within a graphite susceptor which is heated by infrared lamps. The sources are thermally decomposed within the reaction chamber by the elevated susceptor temperatures. The unreacted

Fig. 12.1 Schematic of the Aixtron 200/4 MOCVD system used to grow the QD samples and devices

sources and byproducts are passed through a dry charcoal scrubber before being exhausted to atmosphere.

For nano-scale and quantum dot growth, abrupt interfaces between layers of different composition and precise control on the monolayer (ML) scale are needed. This is achieved through a combination of techniques. Firstly, the schematic in Fig. 12.1 shows two additional lines called the Hydride and MO vent lines. This enables the sources to be vented and the gas flows to stabilise, prior to switching them into the reaction chamber. The differential pressure between the vent and run lines is kept as low as possible, again aiding faster switching. Secondly, the electronically controlled pneumatic valves allow precise and automated control of the gas switching, reproducible to within 0.1 s. Finally, the total reactor gas flow is kept constant (8 slm in our case) by switching on/off so called "Dummy lines" to compensate for the changing number of source flows entering the reaction chamber.

In this chapter the deposition times are specified to an accuracy of 0.1 s, coverage to 0.025 ML, the temperature to 10 °C. We would like to stress that these accuracies are nominal and dependent upon external instrumentation as well as our specific reactor setup. Therefore the values given should be taken as a guide only and more value placed upon the overall trends observed.

12.3 Self-assembled Growth of In(Ga)As/GaAs Quantum Dots Using MOCVD

12.3.1 The Stranski-Krastanow Growth Mode

Initial efforts at quantum dot growth involved patterning, etching and regrowth of quantum well structures, but suffered from poor interface quality and low surface coverage. The major break-through came in 1985 with the discovery that defect free semiconductor quantum dots with strong luminescence could be formed in a self-assembly process via the Stranski-Krastanow growth mode (Goldstein et al. 1985).

There are three different modes in which epitaxial growth can proceed as illustrated in Fig. 12.2. The Frank-Van der Merve mode (Fig. 12.2A) occurs for lattice matched systems and consists of 2 dimensional (2D), layer by layer growth. The second mode is called the Volmer-Weber growth mode (Fig. 12.2B). This takes place for systems with a large lattice mismatch or high interfacial energy, which drives the direct formation of islands. These islands tend to be dislocated and therefore not of use for device applications.

To form device quality In(Ga)As/GaAs quantum dots, the Stranski-Krastanow mode is typically used (Fig. 12.2C). This mode relies on a slight lattice mismatch between the epilayer and substrate, ~7.2% for InAs on GaAs and 3.6% for In_{0.5}Ga_{0.5}As on GaAs, which drives the formation of islands after an initial period of layer by layer growth. Initially the deposited In(Ga)As compresses to fit the smaller GaAs lattice and 2D growth proceeds. However, with each additional layer, the strain energy accumulates until it becomes energetically favorable for islands to

Fig. 12.2 Schematics of the three epitaxial growth modes: (A) Frank-Van der Merve, (B) Volmer-Weber, and (C) Stranski-Krastanow. Reproduced from (Sears 2006)

form. The quantum dots or islands therefore sit on top of a thin 2D layer, called the wetting layer (which is essentially a very thin quantum well). The layer thickness at which the 2D-3D growth transition occurs is called the critical thickness and depends on the lattice mismatch. For the $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ system the critical thickness is 4 monolayers (ML), whereas for the InAs/GaAs system, the critical thickness is much smaller, only 1.7 ML, due to the larger lattice mismatch. For PL measurements and device applications these quantum dots are typically capped with a GaAs layer.

Whilst the driving force for island nucleation is clearly a thermodynamic one, the nucleation process and final QD characteristics are highly dependent on the growth parameters. This is especially the case for metal organic chemical vapor deposition where growth kinetics play a much greater role. In the following sections the key growth parameters, such as coverage, the V/III ratio, growth temperature and growth interruptions will be discussed.

12.3.2 Coverage

The amount of material deposited, or coverage, is one of the most important parameters for QD formation. If too much material is deposited, large coalesced islands

form, which are detrimental for device performance. On the other hand, if too little material is deposited only a low density of islands (or no islands) form. These trends and the Stranski-Krastanow growth mode are illustrated nicely by the atomic force microscopy images in Fig. 12.3. This example is for $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ quantum dots grown using different coverages. At the lowest coverage of 3.3 ML, the critical thickness has not yet been exceeded so that only a thin wetting layer has formed. The monolayer steps can be clearly observed. At 4.7 ML, the critical thickness has only just been exceeded so that a low density ($8 \times 10^9 \text{ cm}^{-2}$) of shallow islands form. These islands have a height of only $\sim 2 \text{ nm}$ and a base diameter of $\sim 50 \text{ nm}$. As more material is deposited, the dot density and height increase up until a saturation point. In this example the density saturates at a coverage of 5.7 ML, at which stage the islands have a density of $\sim 5 \times 10^{10} \text{ cm}^{-2}$, a height of $\sim 5 \text{ nm}$ and a base diameter of $\sim 20 \text{ nm}$. Deposition of further material leads to the formation of large coalesced islands and a reduced island density and height. One large coalesced island can be seen in the lower left hand corner of Fig. 12.3(d). These large coalesced islands are typically dislocated and act as material sinks, growing at the expense of the surrounding islands. Fig. 12.4 shows an extreme example where many of these large coalesced islands have formed.

Fig. 12.3 $1 \times 1 \mu\text{m}^2$ deflection AFM images of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ QDs grown using coverages of (a) 3.3, (b) 4.7, (c) 5.7, and (d) 6 ML. The other growth parameters were kept constant (1.7 ML/s, V/III ratio = 15, 550°C). Adapted from (Tan et al. 2007)

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Fig. 12.4 $1 \times 1 \mu\text{m}^2$ deflection AFM image of an $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ QD sample formed using too much material. A high density of coalesced islands is observed

Figure 12.5 shows PL spectra for the same $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ QD samples discussed above. At 3.3 ML the PL spectrum is centered at 890 nm with a relatively narrow line width, characteristic of a quantum well. As more material is deposited, the photoluminescence shifts to longer wavelengths (red-shifted) as islands form and grow in size. The PL line width also increases substantially which is due to the random nucleation process and resulting large island size distribution. At 8.3 ML the PL intensity drops as more and more large coalesced islands are created.

The effect of coverage on InAs/GaAs QD formation is very similar except that the higher lattice mismatch (7.2%) leads to a much faster nucleation process. This

Fig. 12.5 77 K photoluminescence spectra of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ QDs grown using coverages of 3.3, 4.7, 5.7, 6.0 and 8.3 ML. The other growth parameters were kept constant (1.7 ML/s, V/III ratio = 15, 550 °C). Adapted from (Sears et al. 2006b)

grown using coverages of
kept constant (1.7 ML/s).

Fig. 12.6 Plan view TEM images of InAs/GaAs quantum dots grown using coverages of 1.90, 1.925 and 2.0 ML. The other growth parameters were kept constant (0.25 ML/s, V/III ratio = 40, 520 °C). The QDs are also capped by a 300 nm GaAs layer. Adapted from (Sears et al. 2004)

is illustrated by Figs. 12.6(a), (b) and (c) which show plan-view TEM images of InAs QD samples formed using 1.90, 1.925 and 2.0 ML. In these plan-view TEM images each dot appears as a 'donut' like contrast. At a coverage of 1.90 ML only a low density of islands has formed. However, when the coverage is increased by only 0.025 ML, a high density of islands with minimal defects and strong PL (not shown) results, while a further 0.075 ML leads to dislocations. Clearly the nucleation process is extremely fast in the case of InAs QDs making it a more challenging system to grow. Due to the more challenging nature of the InAs/GaAs QD system, the selective area epitaxy results discussed later in Section 12.6 concentrate on the use of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ QDs. This example also stresses how important it is to avoid the formation of large coalesced islands which are very susceptible to dislocations.

12.3.3 Growth Temperature

Temperature affects the adatom mobility and therefore the density and size of the QDs. Each nucleation site acts as a sink for the surrounding material, essentially creating a depleted zone within which the adatoms are collected and further dot nucleation is inhibited. Therefore, at higher temperatures, a lower density of larger QDs tends to form due to an increased diffusion length and vice versa. These

Fig. 12.7 500 × 500 nm² deflection AFM images of In_{0.5}Ga_{0.5}As/GaAs QDs grown at temperatures of 500, 520, 550 and 570 °C. Adapted from (Tan et al. 2007)

trends are illustrated in Fig. 12.7 which shows AFM images of In_{0.5}Ga_{0.5}As QDs deposited at different temperatures. With increasing growth temperature, the QD height increases from ~1 nm to 5.5 nm and the QD dot density decreases, consistent with an increased adatom mobility. At the highest temperature of 570 °C, the dot density decreases dramatically as large coalesced islands form.

The effects of growth temperature are also nicely illustrated by the PL spectra in Fig. 12.8. With increasing growth temperature, the PL emission shifts to longer wavelengths, characteristic of larger QDs. The PL intensity initially increases, which is likely due to improved material quality. However it then begins to drop at the highest growth temperature which we suspect is due to defects/dislocations forming in the larger QDs. Due to the temperature dependent adatom mobility and cracking efficiency of the precursors (discussed below), the optimum coverage varies with growth temperature. Therefore a study of coverage was made for each QD growth temperature and an optimum temperature of 520 and 550 °C identified for the InAs and In_{0.5}Ga_{0.5}As QDs respectively.

The above studies have concentrated on QD growth temperatures between ~500 and 550 °C. These growth temperatures are relatively low, especially for MOCVD growth and are needed in order to reduce the adatom mobility and hence the tendency for islands to coalesce. However these low growth temperatures also introduce added complexities for QD growth by MOCVD. Temperatures between 600–750 °C are typically used in MOCVD growth to ensure proper cracking of