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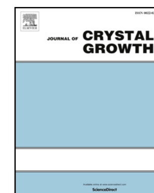

Critical V/III dependence of formation of high density GaSb/GaAs quantum dots grown by AP-MOVPE

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ABSTRACT

The effect of AP-MOVPE growth parameters on the formation of GaSb QD structures on GaAs with TMSb and TEGa as Sb and Ga sources has been studied. Formation of small, coherent GaSb dots on GaAs improved with decreasing V/III ratios which were controlled through changing either TMSb/TEGa at a constant temperature or changing growth temperature at constant TMSb/TEGa ratio. A summary of required source vapour V/III ratios for dot growth at various temperatures is also presented. The maximum effective V/III ratio for dot formation was 0.175. Dot density was more sensitive to growth time as time determines the amount of deposited dot material. Dot density increased with increasing growth time. The shape and size was more sensitive to source vapour mole fraction which controls the growth rate. Lower mole fraction gave smaller sized dots with more uniform distribution compared to higher mole fraction.

1. Introduction

GaSb quantum dots (QDs) in a GaAs matrix have some unique and appealing properties that are being continually exploited. This system has a type-II band alignment, providing strong spatial confinement for holes, while only binding electrons via Coulomb interaction [1,2]. This leads to optical properties that are different from those of type-I QDs, such as a long radiative lifetime, a dot-shape dependent oscillator strength, and large tunability of emitted/absorbed photons. The optical properties of GaSb/GaAs QD structure depend on the physical properties such as dot size shape, composition etc. These physical parameters can in turn be influenced by the growth conditions and parameters such as growth temperature and growth sequence and growth interruption between some of the steps [3,4]. Desired properties for good quality dots is high density of small coherent islands, good size uniformity and minimal (or avoid) formation of dislocated large islands. Molecular beam epitaxy (MBE) is the most widely used technique for fabrication of these structures [3–11]. Just like MBE, metalorganic vapour phase epitaxy (MOVPE) in principle also provides the control over the deposition conditions required to systematically study QD formation.

MOVPE has previously been used to grow GaSb on GaAs [2,12–21]. Low pressure [2,12–15] and atmospheric pressure [16–21] systems have been used with various combinations of organometallic sources. For example, a combination of triethylgallium (TEGa) with triethylantimony (TESb) at low pressure [12,14,15], trimethylgalllium (TMGa) with TESb at low pressure [2,13], and TMGa with trimethylantimony (TMSb) at atmospheric pressure [16–20] have previously been employed. We have recently reported on the fabrication of capped GaSb/GaAs quantum dot structures using an MOVPE system with TEGa and TMSb as Ga and Sb sources, respectively [22] and to the best of our knowledge there is no other published report on the use of a combination of TEGa with TMSb at either low or atmospheric operating reactor pressure for growth of GaSb nanostructures on GaAs.

The aim of this work is to systematically study the effect of growth parameters on the formation of GaSb nanostructures on a GaAs substrate in order to determine the conditions for achieving the desired small-sized, uniformly-shaped and uniformly-distributed nanostructures with a high areal density. Specifically, the study looks at the effect of the TMSb/TEGa (V/III) ratio, growth temperature, total source mole fraction, and growth time in controlling feature size, shape and density.

2. Experimental procedures

Samples in this study were fabricated using a research-scale Thomas Swan MOVPE growth system (Epitor 04) that operates at atmospheric pressure with palladium diffused hydrogen as the carrier gas and a lamp heated molybdenum susceptor. TEGa, TMSb and tertiarybutylarsine (tBAs) were used as Ga, Sb and arsenic sources respectively. The substrates used were semi-insulating (1 0 0) GaAs substrates, misorientated

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by 2° towards (1 1 1)B. Pieces of substrate were blown clean by dry nitrogen after cleaving from the wafer. No further cleaning of the substrates was required as they were bought ‘epi-ready’. Subsequently, they were placed on the susceptor and loaded into the horizontal quartz reactor which was then purged with hydrogen for 10 min. This was followed by annealing at 600 °C for ten minutes in order to remove any surface oxide. The annealing was done with tBAs overpressure in order to prevent substrate surface deterioration. While maintaining a tBAs overpressure the susceptor was set to a chosen GaSb growth temperature. In order to avoid an arsenic background, the reactor was purged for 30 s prior to GaSb dot growth. GaSb dots were grown by simultaneously introducing TEGa and TMSb at a chosen TMSb/TEGa ratio into the reactor for a chosen growth time. After GaSb, the reactor heater was switched off and samples allowed to cool down to room temperature (RT) before being removed for analysis. The morphology of these uncapped dot samples was investigated by scanning probe microscopy (SPM) to study the shape, size and density. A Bruker Dimension FastScan SPM was used for this purpose. Capped QD samples were used for photoluminescence (PL) analysis. In these samples, the dots were ‘cold capped’ with GaAs. This is a two-step process involving the deposition of the first ∼10 nm of the GaAs capping layer at the same temperature used for dot deposition, followed by the deposition of the final ∼70 nm of the capping layer at a higher temperature (650 °C) that is more suitable for obtaining high quality GaAs.

3. Results and discussion

GaSb quantum dots typically form on the surface of GaAs via self-assembly. For this to take place, the surface of GaAs needs to be wetted with GaSb. Due to the lattice mismatch between GaSb and GaAs the GaSb wetting layer has high elastic potential energy. As more GaSb material is deposited on the substrate, additional atoms group together to form the quantum dot in order to reduce this elastic energy. The size and density of the quantum dots depend on the growth conditions. A series of experiments were performed to find suitable conditions for the formation of a high density of dots with an appropriate size and the results presented here are chosen to highlight the key findings and conclusions.

3.1. Dot formation

Fig. 1 shows GaSb structures grown at various temperatures (between 490 °C and 550 °C) on GaAs, using a constant V/III ratio in the vapour phase of 0.5 and a total source mole fraction of 1.58×10^{-4}. Both the lateral size and height of the visible island-like features decrease with a decrease in growth temperature. This is accompanied by an increase in the areal density. For these growth conditions, uniform, small islands begin to appear for a growth temperature of 515 °C and lower. Although the V/III ratio in the vapour phase was kept constant it needs to be pointed out that a decrease in growth temperature will decrease the effective V/III ratio on the growth front, since TEGa is expected to be completely pyrolysed in this growth temperature range [23,24], while the pyrolysis of TMSb reduces with decreasing temperature in the range studied [25]. Therefore, it is highly probable that the trend observed in Fig. 1 is related to a decrease in the effective V/III ratio on the GaSb surface.

In order to test this possibility, a series of experiments was performed where the V/III ratio in the vapour phase was intentionally changed, while the other growth parameters were fixed. Some of the results are displayed in Fig. 2. The surface feature trend in Fig. 2 with varying V/III ratio is similar to that observed in Fig. 1 for a varying growth temperature. Interestingly, the small change in V/III ratio from 0.25 to 0.2 between (c) and (d) caused a significant difference in size, shape and density of the dots.

The above results illustrate that the V/III ratio is the crucial factor in the formation of GaSb quantum dots on GaAs. A reduction in the concentration of Sb-species (compared to Ga-species) on the growth front improves the formation of small, uniform dots with a high areal density. Dot formation is determined by the balance of elastic strain energy, surface free energy and migration of the growth species. The V/III ratio has previously been found to be crucial in the formation of InAs QDs during low pressure MOVPE [26]. For the InAs material system, it was shown that higher V/III ratios yielded smaller, more uniform dots. This was attributed to a reduction in the migration length of the In atom with increasing arsenic partial pressure. It must be noted that in the InAs system, In is the species with the lower mobility. Elemental Sb, on the other hand, is known to have a very low surface mobility during MBE growth of GaSb [27]. Indeed, control of the GaSb dot size and lateral aspect ratio by tailoring the V/III ratio has been used before during MBE growth of GaSb dots. It was illustrated that a lowering of the flux of the immobile species (i.e. Sb) leads to more isotropic and smaller dots [28].

During the formation of GaSb QDs by MOVPE, it is evident from Fig. 2 that a higher concentration of Sb-species on the GaAs surface causes anisotropic lateral growth rates and a lower density of large dots, due to longer migration lengths of the Sb species on the growth front. Also having more of the immobile Sb species on the surface should promote growth around the sites where GaSb nucleation has already occurred, hence the observed increased size (height and lateral size) of the island-like features with increased Sb partial pressure.

GaSb dots were therefore grown on GaAs over the temperature range from 490 °C to 550 °C by “optimizing” the V/III ratio for each growth temperature. Fig. 3 displays the maximum V/III ratio in the vapour phase for the formation of a high density of small dots for the various temperatures chosen in this study. The dashed blue[1] line is an aid to the eye and serves to separate the growth domain leading to a high density of small dots from that leading to a low density of large, elongated dots. Considering the fact that TEGa is expected to be fully pyrolyzed in this entire temperature range of study, while TMSb is fully pyrolyzed only at 550 °C, the effective V/III ratio on the surface at 550 °C should be close to the V/III ratio in the input stream. In this study, the maximum V/III ratio for dot formation at 550 °C (in the MOVPE reactor used) is 0.175. This is lower than what was previously reported for a similar growth system (horizontal AP-MOVPE) but with a different combination of organometallic sources (trimethylgallium (TMGa) and TMSb) and at a slightly lower growth temperature (540 °C) [19]. However it is in the same range (0.2) as the value reported for low pressure MOVPE using TMGa and TESb at 525 °C [2].

3.2. Size and distribution

3.2.1. Growth time

The growth time is expected to control the amount of deposited GaSb and will influence the density of the deposited features [6] as well as possible coalescence of neighbouring features. Under suitable growth conditions for the formation of dots, an increase in the growth time should increase the density and average size of the dots, until neighbouring dots begin to coalesce. This is illustrated in Fig. 4, which shows SPM images of GaSb dots grown at 530 °C with a total source mole fraction of 2.3×10^{-4}, a V/III ratio of 0.2 and for times of (a) 3 s and (b) 4 s, respectively. The dot density for (a) was approximately 300/μm^2, while it was 400/μm^2 for (b). Also, several of the dots (examples encircled in red) show signs of coalescence in (b). Further statistics on these two samples are presented in Table 1. This data was obtained using the ‘particle analysis’ function of the NanoScope analysis software by adjusting the threshold height until a maximum number of dots have been counted. It is clear that the dot size and shape are comparable for both samples, while the sample with the higher growth time had a

[1] For interpretation of color in Figs. 3 and 4, the reader is referred to the web version of this article.

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