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Optimising uniformity of InAs/(InGaAs)/GaAs quantum dots grown by metal organic vapor phase epitaxy

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Abstract

A route towards optimisation of uniformity and density of InAs/(InGaAs)/GaAs quantum dots grown by metal organic vapor phase epitaxy (MOVPE) through successive variations of the growth parameters is reported. It is demonstrated that a key parameter in obtaining a high density of quantum dots is the V/III ratio, a fact which was shown to be valid when either AsH₃ (arsine) or tertiary-butyl-arsine (TBA) were used as group V precursors. Once the optimum V/III ratio was found, the size distribution was further improved by adjusting the nominal thickness of deposited InAs material, resulting in an optimum thickness of 1.8 monolayers of InAs in our case. The number of coalesced dots was minimised by adjusting the growth interruption time to approximately 30 s. Further, the uniformity was improved by increasing the growth temperature from 485 °C to 520 °C. By combining these optimised parameters, i.e. a growth temperature of 520 °C, 1.8 monolayers InAs thickness, 30 s growth stop time and TBA as group V precursor, a full-width-half-maximum (FWHM) of the low temperature luminescence band of 40 meV was achieved, indicating a narrow dot size distribution.

Keywords: Quantum dot; Epitaxy; MOVPE; InAs/GaAs; TBA; Growth

1. Introduction

Over the past 15 years there has been intense research on quantum dots (QDs) and the associated growth conditions [1], [2], [3] and [4], due to their potential for improved optoelectronic components, such as lasers [5], [6] and [7], semiconductor optical amplifiers [8] and quantum dot infrared photodetectors (QDIP) [9] and [10]. Important advantages of QD lasers are reduced threshold currents and higher temperature stability, while the main advantages of QDIPs, when compared to quantum well infrared photodetectors (QWIPs), are reduced dark current and the possibility to detect radiation at normal incidence. These interesting properties follows from the 3D-confinement of carriers in QDs, but in order to fully attain the 3D related advantages, some criteria of the size and density of the QDs have to be fulfilled. Lasers, for example, require a high uniformity and preferably also high density of QDs in order to obtain a high gain. Several research groups have studied the influence of different growth parameters, such as temperature, growth rate and InAs thickness [11], [12], [13] and [14], but the reports to be found in the literature are not conclusive on the parameters crucial for dot size uniformity. In this article a route towards growth of uniform QDs using metal organic vapor phase epitaxy (MOVPE) is presented. Since the optimum growth parameters depend on the actual growth equipment, the way to optimise the parameters is highlighted rather than the exact parameters.

2. Experimental

The structures used in this study were grown by MOVPE in an Emcore (D-180 LDM) reactor operating at 100 mbar, using the organometallics TEGa, TMIIn and AsH₃ as source materials. Before growth, the GaAs substrate was deoxidised for 5 min at 730 °C and a GaAs buffer layer of 3000 Å was grown at 710 °C. The InAs QDs were grown on a 20 Å thick In_{0.15}Ga_{0.85}As buffer layer. The growth rate for the buffer layer and the QDs were kept fixed at 6.7 Å/s and 0.46 monolayers/s (ML/s), respectively, whereas the V/III ratio, nominal thickness and temperature during growth of the QDs have been varied. After the QD deposition a growth interruption (GI) was applied, during which all precursors were switched off. In this study, three different GI times were tested: 12 s, 30 s and 60 s.

In the last step of the optimisation, two samples have been grown, using tertiary-butyl-arsine (TBA) as precursor instead of AsH₃, omitting the In_{0.15}Ga_{0.85}As buffer layer. The optimised growth parameters from the AsH₃ study (i.e. InAs thickness: 1.8 ML, growth temperature: 520 °C, GI: 30 s) have been employed, while the V/III ratio was varied. These samples have been capped by 350 Å GaAs, which was grown at a growth rate of 3 Å/s, while the temperature was ramped up to 600 °C. Thereafter, a second QD layer was grown at 520 °C intended for characterisation with atomic force microscopy (AFM).

AFM was used to measure the topography of the dots. Statistical information, such as the dot density, was obtained from the AFM images. The photoluminescence (PL) measurement was performed at 10 K using a continuous flow helium cryostat, an argon ion laser (514.5 nm) and a LN₂-cooled Ge-detector.

3. Quantum dot self assembly

QDs are grown using self assembly in the Stranski–Krastanow growth mode [15] and [16], which is an energy driven process, where the system minimises energy through transition from a highly strained layer (2D) to coherent islands (3D). There are four different phases for

the QD formation in this 2D to 3D transition: 2D layer-by-layer growth, nucleation, island growth and ripening [16]. In order to grow uniformly distributed and size homogenous QDs, the four growth phases must be optimised. The nucleation phase, which is the crucial step for the density of dots, can be influenced by different growth conditions, such as temperature, growth rate and V/III ratio. The amount of deposited material affects the extension of the island growth phase, while the length and conditions during the ripening phase can influence the size distribution.

4. Results and discussion

4.1. Influence of the V/III ratio on the QD density

Growth parameters during the QD growth were varied in a systematic manner in order to attain a homogenous QD ensemble. The first parameter considered was the V/III ratio. In Fig. 1a–c, the variations of the QD density, n , can be seen, where the V/III ratio was reduced successively from 33 ($n = 8E9 \text{ cm}^{-2}$) to 12.7 ($n = 9.7E10 \text{ cm}^{-2}$) and finally to 6.2 ($n = 7.8E10 \text{ cm}^{-2}$). The density variations can be explained by differences in the migration length, caused by different V/III ratios. At high V/III ratios the density of As adatoms on the growth surface is high, which effectively decreases the migration length of In-species before incorporation into the crystal. Conversely, at low V/III ratios the migration length is increased, which tends to enhance any energy-minimising rearrangements of the growing layer into a more 3D-like topology [13], [17] and [18]. Such generated local thickness variations will provide nucleation sites for the QDs. In Fig. 1b, the V/III ratio gives a migration length of the In-species that is sufficiently long to produce effective nucleation sites, yet short enough to maintain a high density of these sites. Halving the V/III ratio as in Fig. 1c leads to an increased migration length, and thus to a somewhat reduced dot density. By contrast, in Fig. 1a the migration length is probably too short to produce suitable nucleation sites at all—the wetting layer is essentially 2D throughout the deposition phase. Due to the lack of local thickness variations there will be no preferred nucleation sites, which might explain the low density of dots. Furthermore, since the metastable phase is prolonged, the wetting layer will be thicker before entering the nucleation phase, which gives rise to larger non-coherent islands [16]. The V/III ratio that produced the highest dot density (V/III = 12.7) was chosen for further improvement of the uniformity.

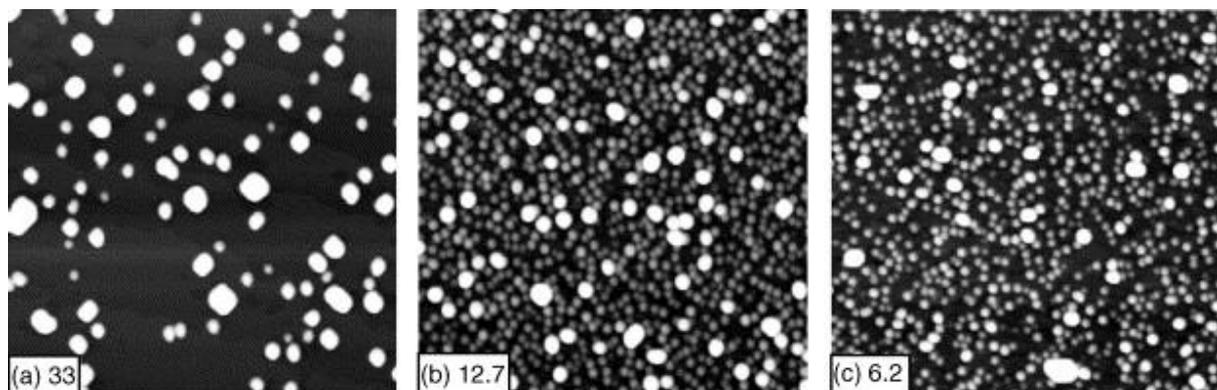


Fig. 1. AFM images ($1 \mu\text{m} \times 1 \mu\text{m}$) of QDs grown with different V/III ratios: (a) 33, (b) 12.7, (c) 6.2. Growth rate: 0.46 ML/s, InAs thickness: 2.3 ML and GI: 12 s.

4.2. Influence of the InAs thickness on the uniformity

The next step was to optimise the amount of material deposited, in order to remove the large dots seen in Fig. 1b. Fig. 2a–c shows that the number of large dots varies with the nominal thickness of deposited InAs. When the InAs thickness was decreased successively from 2.3 ML to 1.8 ML, the number of large dots was reduced from $1\text{E}10\text{ cm}^{-2}$ to $3.8\text{E}9\text{ cm}^{-2}$, while the total density increased from $9.7\text{E}10\text{ cm}^{-2}$ to $1.15\text{E}11\text{ cm}^{-2}$. A further reduction of the InAs thickness from 1.8 ML to 1.6 ML caused a decrease of the dot density by 22% and an increase in the density of large dots by 68%. Consequently, 1.8 ML was chosen as the optimum thickness. The increased uniformity with decreased amount of deposited material can be explained by the growth evolution of the dots [19]. In the early island growth phase, small dots are formed, corresponding to a low facet angle. If the deposition of material continues after a certain size is reached, the small dots develop into larger dots with a steeper facet angle. It is therefore of great importance to keep the amount of deposited material low, in order to obtain a homogenous dot ensemble.

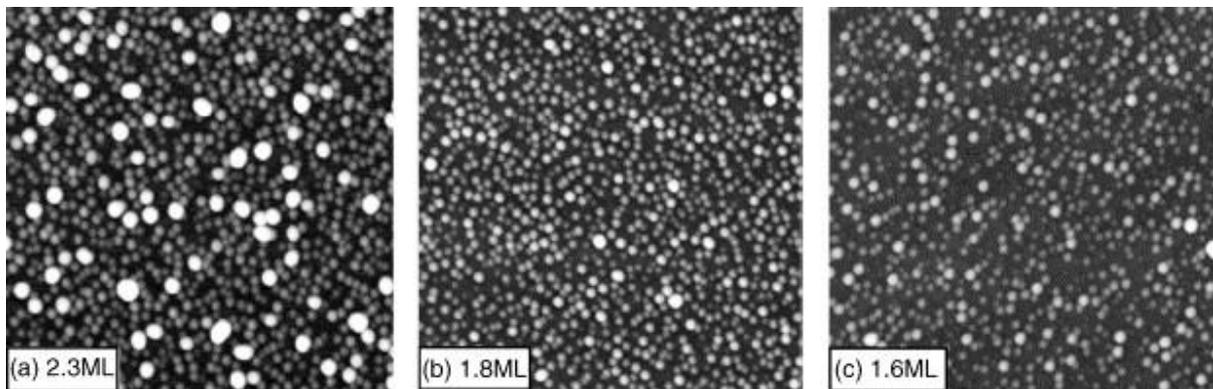


Fig. 2. AFM images ($1\text{ }\mu\text{m} \times 1\text{ }\mu\text{m}$) of QDs grown with a nominal InAs thickness of: (a) 2.3 ML, (b) 1.8 ML and (c) 1.6 ML. Growth rate: 0.46 ML/s, V/III ratio: 12.7 and GI: 12 s.

4.3. Influence of the growth interruption time on the uniformity

So far improvements in the density and uniformity were accomplished by optimising the V/III ratio and decreasing the amount of deposited material, respectively. Nevertheless, the sample still suffered from a number of large coalesced dots (as can be seen in the $3\text{ }\mu\text{m} \times 3\text{ }\mu\text{m}$ AFM image shown in Fig. 3a). To remove these coalesced dots, the duration of the growth interruption (GI) was adjusted. During the GI, surface adatoms from larger coalesced dots migrate towards crystalline QDs, whereby the number of coalesced dots decreases and the average dot size increases [4] and [17]. Fig. 3b shows how the number of coalesced dots varies with the GI times chosen (12 s, 30 s and 60 s). As the GI time is extended from 12 s to 30 s a reduction of the number of coalesced dots occurs, while the extension of interruption time from 30 s to 60 s results in a marginal deterioration in the number of coalesced dots. Accordingly a GI time of 30 s was chosen. The GI time has an obvious influence on the dot height (see Fig. 3b) as well as the lateral dimensions. The average dot diameter increased from 15 nm to 18 nm, when the GI was prolonged from 12 s to 30 s and to 21 nm when a 60 s GI was employed.

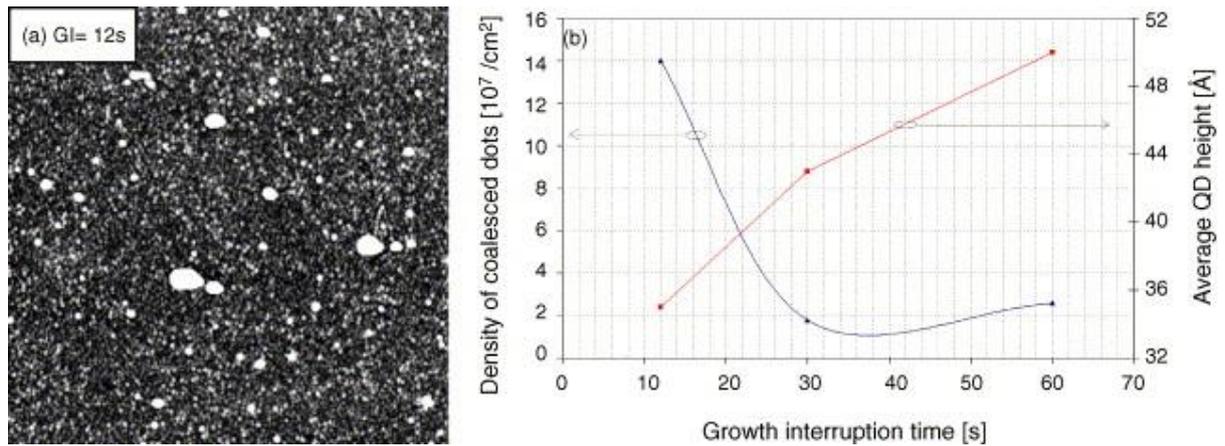


Fig. 3. (a) AFM image ($3 \mu\text{m} \times 3 \mu\text{m}$) of QDs grown with a GI time of 12 s. (b) Influence of the GI time on the number of coalesced dots and the dot height, respectively. Growth rate: 0.46 ML/s, V/III ratio: 12.7 and InAs thickness: 1.8 ML.

4.4. Influence of growth temperature on the uniformity

The dot uniformity was further optimised by increasing the growth temperature from 485 °C to 520 °C (Fig. 4), i.e. the size deviation from the average dot size dropped from 35% to 20%. As a consequence of the temperature increase, the dot density decreased to $3 \times 10^{10} \text{ cm}^{-2}$ and the mean dot height and diameter increased to 6 nm and 24 nm, respectively. This is due to a higher growth rate of the islands at higher temperature and an associated higher mobility of atoms on the surface. When the consumption rate of excess material in the wetting layer is high, a limited number of islands are formed before the strain energy in the wetting layer has dropped below the activation energy needed for nucleation.

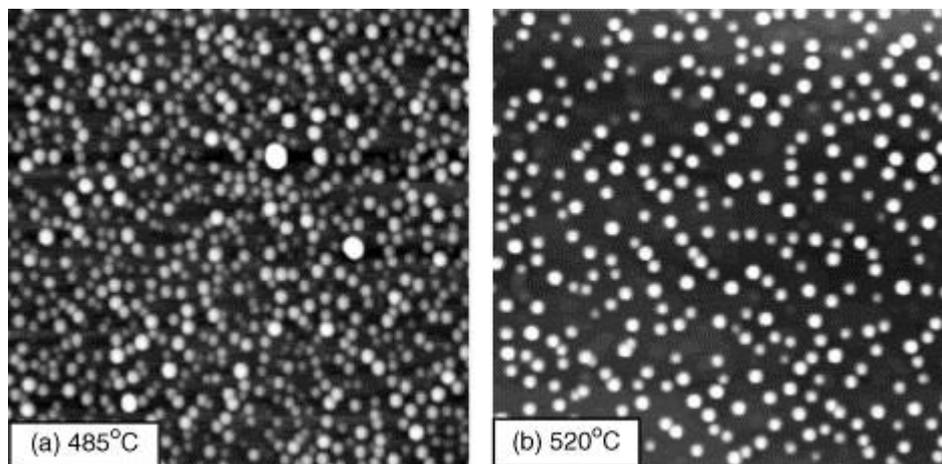


Fig. 4. AFM images ($1 \mu\text{m} \times 1 \mu\text{m}$) of QDs grown at a temperature of: (a) 485 °C and (b) 520 °C. Growth rate: 0.46 ML/s, V/III ratio: 12.7, InAs thickness: 1.8 ML and GI: 30 s.

4.5. Effect of TBA as precursor

As a next step in the route towards a homogenous dot ensemble, TBA was employed as group V precursor instead of AsH_3 . Since the V/III ratio was identified as the most important parameter to increase the density of dots from the previous results, the starting point was to vary this parameter, while keeping the optimised growth parameters for the InAs thickness

(1.8 ML), temperature (520 °C) and GI (30 s). Due to the higher cracking efficiency of TBA relatively AsH_3 at these temperatures, an initial V/III ratio of two was chosen (Fig. 5a). Large coalesced dots were formed at this V/III ratio, indicating a too high V/III ratio for good growth conditions. By reducing the V/III ratio by a factor of two (Fig. 5b), a relevant regime for growth of QDs was found, where the average height and diameter of the dots were 5.5 nm and 25 nm, respectively, with a deviation from the average dot size of 15%. The density is three times smaller than for the corresponding sample grown with AsH_3 , which is attributed to the removal of the InGaAs buffer layer [20]. The luminescence spectra (Fig. 6) of this sample reveal two peaks marked E_0 and E_1 , corresponding to the interband transitions between the two lowest quantised energy levels of the dots. A FWHM of 40 meV of the ground state transition (E_0), indicates a relatively homogenous dot ensemble, which could be further improved by tuning of the growth parameters, according to the route employed earlier. This shows that the route towards optimisation used is consistent for both AsH_3 and TBA, with a resulting high uniformity of QDs.

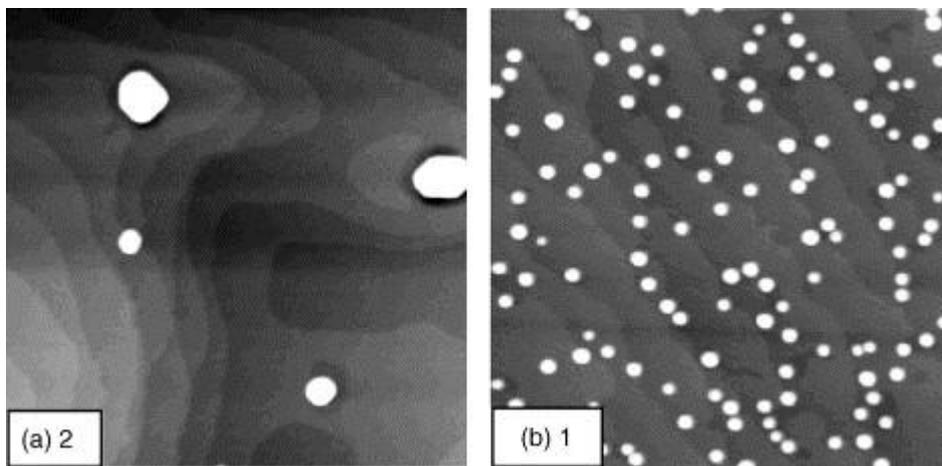


Fig. 5. AFM images ($1 \mu\text{m} \times 1 \mu\text{m}$) of QDs with different V/III ratios: (a) 2 and (b) 1, when using TBA as precursor. Temperature: 520 °C, growth rate: 0.46 ML/s, InAs thickness: 1.8 ML and GI: 30 s.

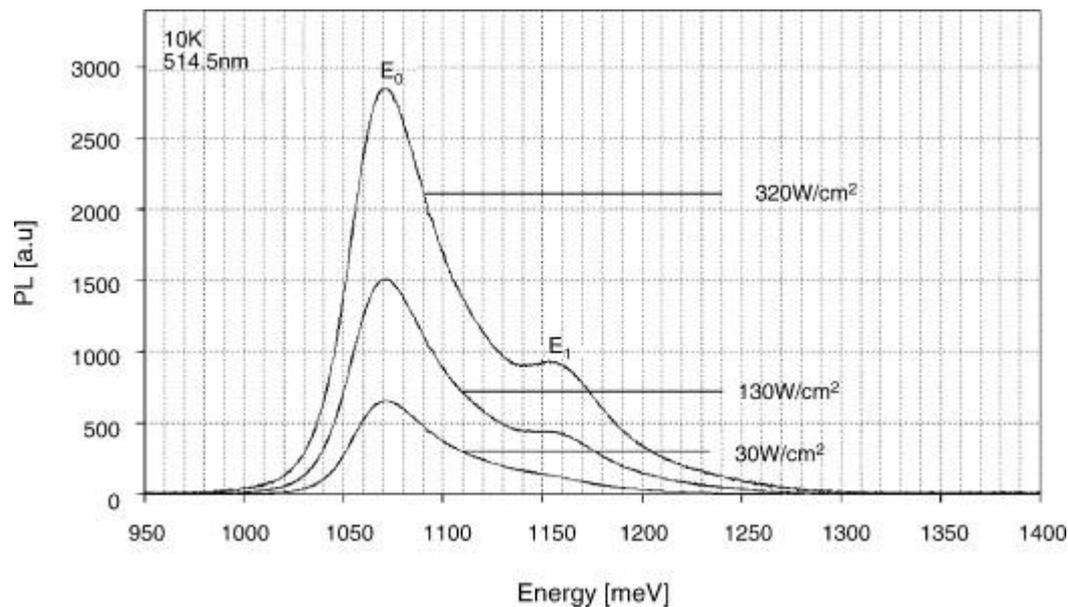


Fig. 6. Photoluminescence measurements at 10 K for varying excitation power, revealing the interband transitions E_0 (ground state) and E_1 (excited state) of the QDs in Fig. 5b.

5. Conclusions

By a systematic variation of growth parameters the uniformity of QDs has been improved. The V/III ratio was shown to be crucial for the density of dots, while the nominal InAs thickness and growth interruption served to improve uniformity. By increasing the temperature, the uniformity increased, but simultaneously the dot density decreased. Through a combination of the optimised growth parameters and by using TBA as precursor, a high uniformity of InAs QDs has been achieved.

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