InAs Quantum Dots Grown by MOCVD for Mid-infrared Emission

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ABSTRACT

Mid-infrared InAs QDs grown on In$_x$Ga$_{1-x}$As/InP matrix by low pressure metal organic chemical vapor deposition have been studied. Formation of the InAs QDs with different growth conditions has been investigated. It has been found morphology of the InAs QDs on such In$_x$Ga$_{1-x}$As/InP matrix is very sensitive to the growth conditions. InAs QDs with density of 1.3×10$^{10}$ cm$^{-2}$ were grown by using S-K growth method. To improve the dot size uniformity, a two-step growth method has been used. Using the two-step growth method, the InAs QDs size uniformity has been improved by 63% and 110%, respectively, when compared that of the dots grown by ordinary S-K method and ALE method. FWHM of the PL curve at 77K of the QDs grown by using the two-step growth method was measured of 26 meV and the peak emission wavelength was at >2.3µm.

Keywords: Quantum dots, MOCVD, mid-infrared

1 INTRODUCTION

Recently, attempts to use quantum dots (QDs) for developing high performance mid-infrared (IR), 2-4 µm wavelength range, lasers have attracted much interest [1-3], because mid-IR lasers are attractive for application in analysis of pollutant and combustible gases, remote sensing and ranging, as well as the medical lasering surgery, etc. However, to the best of our knowledge, the longest emission wavelength from common InAs QD structure reported so far only reaches near 2 µm at room temperature. Several research groups used InAs-rich ternary alloys, such as InGaAs[4], InAlAs[5] and InAsP[6] to form QDs with larger dot size by reducing the lattice mismatch between the QD layers and InP substrate so that to extend the QDs’ emission wavelength. The longest light emission wavelength from these QDs structures was still limited at about 2 µm. By employing the lowest energy bandgap binary and ternary in conventional III-V semiconductor alloys, InSb and InAsSb, to form QDs on InP substrate, the emission wavelength was pushed to 2.2 µm [7,8].

In this paper, we reported the extension of light emission wavelength of InAs QDs longer than 2 µm by embedding the InAs QDs in In$_x$Ga$_{0.1-x}$As graded matrix layers.

2 EXPERIMENTS

All the samples were grown in an horizontal MOCVD reactor in Aixtron system. Low toxic MO sources tertiarybutylarsine (TBA) and tertiarybutylphosphine (TBP) were used as the group V precursors in the MOCVD growths. While trimethylindium (TMIn) and trimethylgallium (TMGa) were used as the group III sources. Purified N$_2$ was used as the carrier gas. The MOCVD reactor pressure was set at 20 mbar for InAs QD growth, while for growing the other layers it was set at 100 mbar. On InP (001) substrate, a 200 nm InP buffer was grown at 600-630 ºC after thermal annealed the substrate at 650 ºC for 8 min. To extend the emission wavelength, the InAs QDs were embedded in the indium graded In$_x$Ga$_{1-x}$As layers. The indium content of the graded In$_x$Ga$_{1-x}$As matrix layer of the QD structure was linearly increased from x = 0.53 (lattice matched to InP) to x = 0.65 ~ 0.80. The graded In$_{0.53-y}$Ga$_{0.47+y}$As layers were grown at 600 ºC. Thickness of the In$_{0.53-y}$Ga$_{0.47+y}$As graded matrix layer was 30nm and its indium content was increased linearly from 0.53 to a target composition with different gradations y of 0, 0.12, 0.19 and 0.27, respectively. After that, the substrate temperature was lowered down to 550 ºC for growing the InAs QDs. Morphology of the QDs was measured using an atomic force microscope (AFM). The PL measurements were performed by using a 532 nm diode laser as the exciting source. The PL signal from the sample was detected by a cooled PbS photodetector. Standard lock-in technique was used to amplify the signal before sent it to PC for processing.

3 RESULTS AND DISCUSSION

3.1 Morphology of QDs
The morphology and dot density of the InAs QDs of the samples were measured by atomic force microscopy (AFM). Figure 1 shows the 1×1 μm AFM images of the InAs QDs grown by using different growth methods.

![AFM images of InAs QDs](image)

Figure 1: 1×1μm AFM top mages of sample (a), (b) and (c), grown by conventional S-K fast growth, ALE and two-step growth (fast growth + ALE), respectively. Dot lateral size and height distributions are shown in the histograms.

Sample (a) was grown by conventional S-K method with fast growth rate, where the growth rate was 0.83 ML/s. Sample (b) was grown by using atomic layer epitaxy growth with 8 repeated cycles. In each cycle, 40sccm TMIn source flow was opened for 2 second, followed by N₂ purge 1 second and then 50sccm TBA flowed for 3 seconds. For sample (c), a two-step growth [9] was performed with fast growth and combined with the ALE method. The worst uniformity of dots formed comes from the ALE grown sample (b). Sample (b) has the largest dot size fluctuation which is defined as the largest diameter difference between the QDs divided by the mean dot diameter and the largest standard deviation of the QDs’ size. The QDs formed on sample (b) are with dots and dashes. The QD density of sample (b) is as low as 0.9×10¹⁰/cm². The QD density of sample (a) grown by conventional S-K method with fast growth rate reaches 1.3×10¹⁰/cm² with almost no dash found, but the dots formed are with large size dispersion. For sample (c) grown by the two-step (fast growth + ALE), the dot density formed is as high as 1.3×10¹⁰/cm² and at the sample time the dot size uniformity has also been improved much.

For one-step S-K fast growth of QDs, the nucleation rate is high when the deposited InAs wetting layer is thicker than its critical layer thickness because the indium atoms are forced to coalesce, in this case, due to the short surface diffusion time/length, which suppresses forming the dashes. At the same time, the short diffusion leads to form the nuclei with higher density but un-uniform [10]. It has been reported that during the post-growth interruption, the reactor system was in a thermodynamic equilibrium growth, the edge barrier of the formed islands self-limited the further growth [11]. Although this growth can improve the QD size uniformity, the growth is mainly by the diffusion of surface indium atoms. These atoms are limited. As a result, the finally formed QDs by one-step fast growth are still in large size dispersion [12].

It can be observed there are three types of QDs are formed in ALE growth of sample (b). They are anisotropic nanodash along [\(\bar{1}10\)] direction as shown in Fig. 1(b); dashes do not along [\(\bar{1}10\)] direction and dots. In ALE growth fo InAs QDs, TMIn flux was set at low flow rate of 40 sccm, the diffusion length of adatoms is large and has advantages in the anisotropic surface diffusion of In atoms along the [\(\bar{1}10\)] direction, which resulted in forming the dashes along the [\(\bar{1}10\)] direction as shown in Fig. 2(b).

Formation of the dashes not along [\(\bar{1}10\)] direction is caused by the unstable composition or strain of the graded In₀.₅₃₋₀.₇₂Ga₀.₄₇₋₀.₂₈As matrix layer. The InAs dots are formed between the nano-dashes in the ALE growth of InAs QDs. Under the ALE growth, indium and arsenic atoms were alternatively put into the reactor. Because of the edge barrier of the formed QDs, not all the indium atoms diffused to the dash matrix. Some surface atoms cannot reach the formed dashes. They will stay between the dashes and form into dots. Since there are dashes and dots in sample (b), the QDs’ size uniformity is very bad and with low QD density.

Sample (c) was grown by using the two-step method in which the InAs QDs were grown with an initial fast growth + the ALE growth. The fast growth in step 1 growth has two advantages: suppressing the formation of dashes [13] and creating nuclei with high density after the InAs wetting layer growth. In step 2 ALE growth, larger dots formed grow slow while the dots with smaller size grow fast, so to improve the QDs size uniformity. The fluctuation of dots’ lateral size and height of sample (c) is 0.52 and 1.01, respectively, which is the smallest among the three samples.

3.2 Extension of the emission wavelength

Figure 2 shows the 77 K normalized PL spectrum of the three InAs QDs samples. In sample C, InAs QDs are embedded between symmetric In₀.₇₂₋₀.₅₃Ga₀.₂₈₋₀.₄₇As graded
layers; in sample D, the InAs QDs are embedded in the lattice matched \(\text{In}_{0.53}\text{Ga}_{0.47}\text{As}\) barriers; while for sample E, the InAs QDs are embedded in lower bandgap \(\text{In}_{0.72}\text{Ga}_{0.28}\text{As}\) barrier layers. The measured emission wavelength of sample C, D and E is 2.14 \(\mu\text{m}\), 1.96 \(\mu\text{m}\) and 2.28 \(\mu\text{m}\), respectively. The emission wavelength of QDs structure is determined by the bandgap of the QDs material, height of QDs [4,14], the energy bandgap of the barrier [15,16] and the strain of the cap layer [17,18]. The mean dot heights of the three samples as indicated in table 1 are all around 9 nm. Large red-shift of the emission wavelength, \(\geq 220 \text{ nm}\), of sample C and E has been obtained when compared with that of the sample D of QDs of which is embedded in lattice match \(\text{In}_{0.53}\text{Ga}_{0.47}\text{As}\) barriers. The red shift of the samples’ emission peaks is mainly because of their barrier confinement and the cap layer’s strain. Sample E has the lowest barrier height and smallest strain of the \(\text{In}_{0.72}\text{Ga}_{0.28}\text{As}\) cap layer. In sample C, the InAs QDs are embedded in symmetric \(\text{In}_{x}\text{Ga}_{1-x}\text{As}\) graded barriers. It’s cap layer has less strain when compared with that of sample D. At 77K, the measured PL peaks of sample C and E were \(\geq 2.2 \mu\text{m}\). This is the longest emission wavelength of InAs QDs reported so far, to our best knowledge. Figure 3 plots the PL emission intensity and full width of half maximum (FWHM) of the three QD structures. It shows that the InAs QDs formed on graded \(\text{In}_{x}\text{Ga}_{1-x}\text{As}\) barrier has similarly high PL emission intensity and the FWHM to that of the InAs QDs embedded in high energy bandgap lattice-matched \(\text{In}_{0.53}\text{Ga}_{0.47}\text{As}\) barriers. Sample E, in which the InAs QDs is embedded between the low energy bandgap \(\text{In}_{0.72}\text{Ga}_{0.28}\text{As}\) barriers, has the narrowest FWHM PL spectrum of 120 nm, but its emission intensity is the lowest. The narrower PL spectrum of sample E is attributed to its more uniform dot size. Its lower emission intensity is because of the lower energy barrier height. The confinement of the carriers in the QDs is weaker in sample E.

### Table 1 measured QD parameters of the samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sample (C)</th>
<th>Sample (D)</th>
<th>Sample (E)</th>
</tr>
</thead>
<tbody>
<tr>
<td>QD density(cm(^{-2}))</td>
<td>(1 \times 10^{10})</td>
<td>(1.25 \times 10^{10})</td>
<td>(1.32 \times 10^{10})</td>
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<tr>
<td>Mean height (h)</td>
<td>9.0nm</td>
<td>8.9nm</td>
<td>8.9nm</td>
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<tr>
<td>Diameter (D)</td>
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<td>54.0nm</td>
<td>52nm</td>
</tr>
<tr>
<td>(\Delta h) (nm)</td>
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<td>1.17</td>
<td>1.15</td>
</tr>
<tr>
<td>(\Delta D) (nm)</td>
<td>1.09</td>
<td>1.07</td>
<td>1.06</td>
</tr>
</tbody>
</table>

### 4 SUMMARY

High quality InAs QDs have been grown on InP substrate by MOCVD using TBA and TBP as group V sources in pure nitrogen ambient. The QD density reaches \(1.3 \times 10^{10}/\text{cm}^2\) with greatly optimized size uniformity. By using a two-step (fast growth + ALE) growth, the QD density and size uniformity has been improved much. The emission wavelength of InAs QDs is red-shifted by grown the QDs on \(\text{In}_{x}\text{Ga}_{1-x}\text{As}\) matrix layers. Compared with the QDs grown on \(\text{In}_{0.53}\text{Ga}_{0.47}\text{As}\) lattice matched matrix, those grown on strained \(\text{In}_{x}\text{Ga}_{1-x}\text{As}\) matrix have higher QD density. PL emission wavelength of InAs/In\(_x\text{Ga}_{1-x}\text{As}/\text{InP}\) QD structure has been red-shifted from 180 nm to 320 nm, as compared with that of InAs/In\(_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}\) QDs. The longest wavelength, reported so far, of 2.28 \(\mu\text{m}\) at 77 K with narrow FWHM of 26 meV from InAs QDs is obtained.

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