InAs quantum dots grown on InGaAs buffer layers by metal–organic chemical vapor deposition

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Abstract

InAs quantum dots were deposited onto GaAs or onto a thin (nominal 7 ML) In$_x$Ga$_{1-x}$As layer ($x = 0.075$ or 0.15) by metal–organic chemical vapor deposition and compared using photoluminescence measurements, plan-view transmission electron microscopy (TEM) and atomic force microscopy (AFM). The photoluminescence intensity was considerably reduced for samples grown using an InGaAs buffer layer. This is correlated with the formation of dislocations (density $= 1.5 (\pm 1) \times 10^8$ cm$^{-2}$) making them unsuitable for incorporation into devices requiring high optical efficiency.

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1. Introduction

Quantum dot (QD) optoelectronic devices such as infrared detectors and diode lasers offer many improvements over their quantum well counterparts due to their three-dimensional carrier confinement and discrete energy states. InAs/GaAs self-assembled QDs are commonly grown by the Stranski–Krastanow growth mode [1–3]. However this growth mode typically results in a low QD surface coverage (≈10%) and a broad QD size distribution which causes problems such as gain saturation in diode lasers. One approach used to minimize this problem is the insertion of the QDs within a thin In$_x$Ga$_{1-x}$As ($x = 0.1–0.2$) quantum well. This has been shown to improve laser
performance through increased carrier capture by the QDs, resulting in ground state lasing and reduced threshold current density [1,4,5]. Deposition of InAs onto a thin InGaAs buffer layer has also been shown to increase the QD density [1,4,6].

Most studies of quantum dots inserted into an InGaAs quantum well have involved structures grown by molecular beam epitaxy (MBE). In this letter we use metal–organic chemical vapor deposition (MOCVD) to deposit InAs QDs onto InₙGa₁₋ₙAs buffer layers (ₙ = 0, 0.075 or 0.15). These are characterized using atomic force microscopy (AFM), plan-view transmission electron microscopy (TEM) and photoluminescence (PL). In contrast to MBE growth, we found that an InGaAs buffer layer leads to dislocation formation, degrading the QD optical efficiency. We suggest that this is due to local variation in indium segregation from the buffer layer leading to indium enriched QDs and/or the increased strain contributed to the system by the InGaAs buffer in combination with the higher growth temperatures typical of MOCVD.

2. Experimental procedure

The samples studied in this work were grown on (001) S.I. GaAs by low-pressure (100 mbar) MOCVD. Three samples were grown: one with InAs QDs deposited directly onto GaAs and the other two with InAs QDs deposited onto an InₓGa₁₋ₓAs buffer layer with ₙ = 0.075 and 0.15, respectively. The sources used were trimethylindium, trimethylgallium and AsH₃ with H₂ as the carrier gas. The basic structure is shown in Fig. 1 and consists of a buried QD layer for PL and TEM studies and a layer of uncapped QDs on the surface for AFM. A 300 nm GaAs buffer layer was first grown at 650°C. The growth was then interrupted and the sample allowed to cool to 520°C for deposition of a nominally 7 ML InₓGa₁₋ₓAs buffer layer (ₙ = 0.075 or 0.15), followed by InAs to form the QDs. For QDs grown directly on GaAs, the InGaAs buffer layer was omitted. While MOCVD conventionally uses higher (650–750°C) growth temperatures, much lower growth temperatures (450–550°C) are typical for QD growth where it is necessary to reduce adatom mobility and the tendency for the QDs to coalesce [7,2,3]. For device purposes, many stacked layers of QDs are typically needed. Therefore we initially concentrated on InGaAs buffer layers grown at the same temperature as the QDs in the hope that we could avoid temperature ramping during future stacked QD layer growth. However the effect of the InGaAs growth temperature will also be briefly discussed later. The QDs were capped with 300 nm of GaAs. The first 30 nm of this was grown at the same temperature as the InAs QDs and the remaining 270 nm was grown as the temperature was ramped to 600°C. Growth was interrupted again and the sample allowed to cool to 520°C before depositing a final layer of uncapped QDs.

Identical QD growth conditions, except for deposition time, were used for each sample. The InAs deposition time was optimized for each sample to give the best PL intensity. QDs are very sensitive to the amount of material deposited. Too little material results in a low density of smaller QDs leading to low-intensity, blue-shifted PL. On the other hand, if too much material is deposited, large QDs may form and are more susceptible to dislocation/defect formation, again reducing the PL intensity. The optimized InAs deposition time was found to decrease with increasing indium content in the InₓGa₁₋ₓAs buffer layer from 3.8 to 3.4 to 3.0 s for ₙ = 0, 0.075 and 0.15, respectively. This may be explained by (a) the additional strain energy contributed to the system by the InGaAs

![Fig. 1. Schematic of the grown structure. For InAs QDs deposited onto GaAs the InₓGa₁₋ₓAs layer was omitted.](image-url)
buffer layer or (b) indium segregation (from the InGaAs buffer layer to the surface) which plays an important role in determining the onset of islanding [8, 7]. While the InGaAs surface morphology is very different to that of the GaAs buffer (not shown), this is not believed to be responsible for the reduction in deposition time or the reduced PL radiative efficiency (discussed below). InAs QDs were also deposited onto an InGaAs buffer layer grown at 650 °C. At 650 °C we observed that the In_{0.15}Ga_{0.85}As buffer layer has a very similar surface morphology to that of GaAs (not shown). However the deposition time still had to be reduced even further to 2.9 s (from 3.8 s for GaAs) to optimize the PL intensity. As shown later, PL measurements of all these samples show very similar PL emission and linewidth suggesting that structurally they are similar despite the change in deposition time. Plan-view TEM of the buried QDs also show them to be of similar diameter.

Room temperature PL measurements were performed by exciting the samples with a frequency-doubled 532 nm diode-pumped solid-state laser source. The luminescence was dispersed through a 0.5 m monochromator and collected with an InGaAs detector. The buried QD layers were also examined using plan-view TEM. In most cases the surface QDs were etched prior to TEM sample preparation, leaving only the buried QD layer for imaging. Samples for plan-view TEM were prepared by mechanically polishing the samples to 150 μm, followed by dimpling to a thickness of 40 μm and chemically etching with H₂SO₄:H₂O₂:H₂O (3:1:1). TEM analysis was carried out using a Philips CM 300 electron microscope instrument operated at 200 kV.

3. Results and discussion

Figs. 2(a) and (b) are on-zone, bright field, plan-view TEM images of the buried InAs QD layer. The QDs in Fig. 2(a) are deposited directly onto a GaAs buffer and those in (b) onto a 7ML In_{0.075}Ga_{0.925}As buffer layer. As can be seen, both samples show similar dot contrast. Care must be taken in interpreting the dot shape from this contrast because it is largely determined by the strain field around the dots rather than their actual size or shape [9]. For all three samples, the QD density is ≈3.3 × 10¹⁰ cm⁻². Most MBE literature reports increased QD densities for QDs deposited onto InGaAs buffers [1, 4]. However we find that such an increase was accompanied by a reduced
radiative efficiency and that the QD deposition time (and hence density) had to be reduced to maximize the PL intensity. Only a couple of MBE papers report a similar trend [10,11,6].

Fig. 3 shows room temperature PL for InAs QDs grown onto different InGaAs buffer layer compositions (using their optimized deposition time). It is clear that increasing indium in the buffer layer leads to a drop in PL intensity while the full-width at half-maximum (FWHM) and peak wavelength remain relatively unchanged. In spite of some variability in the peak PL intensity, samples grown on an InGaAs buffer layer consistently showed lower PL intensity than those grown on GaAs. A similarly reduced PL intensity was observed for QDs deposited onto a 650°C grown InGaAs buffer. Plan-view TEM images clearly show a high dislocation density for samples grown using an InGaAs buffer layer. Fig. 4(a) is a low magnification, plan-view TEM micrograph of QDs grown on an In$_{0.075}$Ga$_{0.925}$As buffer layer and reveals many dislocations. The dislocation density is $\approx 1.5(\pm 1) \times 10^8$ cm$^{-2}$ and a similar dislocation density is observed for the sample with QDs deposited onto an In$_{0.15}$Ga$_{0.85}$As buffer layer (not shown). Due to the limited image size, dislocation densities could not be calculated more accurately and significant changes in density between the two samples with different InGaAs buffer layers could not be discerned. No such defects were observed for the QDs deposited directly onto GaAs. These defects are potentially responsible for the degraded PL. The dislocations present faster non-radiative recombination paths and compete with the QDs for carrier capture hence reducing the PL intensity. Local variation in indium segregation from the InGaAs buffer layer may be leading to indium enriched QDs which are more susceptible to dislocation formation [12]. Although the InGaAs buffer layers used are well below their known critical thickness, it may also be possible that the combined strain accumulated in the QDs and buffer layer is sufficient to favor dislocation formation, especially during the high-temperature (600°C) growth of the GaAs cap which may promote strain relaxation through dislocation formation.

![Fig. 3. Room temperature photoluminescence spectra for InAs QDs deposited directly onto a GaAs buffer and onto a nominally 7 ML In$_x$Ga$_{1-x}$As buffer layer ($x = 0.075$ or 0.15).](image)

![Fig. 4. Plan-view TEM images taken under on-zone, [001], bright field conditions of InAs dots deposited onto a nominally 7 ML In$_{0.075}$Ga$_{0.925}$As buffer layer. (a) is a low magnification image showing a high density of ripened dots and dislocations. One of these ripened dots and dislocation is shown in figure (b).](image)
The observed dislocations predominantly show an alignment close to one of the \(\langle 110\rangle\) directions. Figs. 4(a) and (b) have the same orientation and a large number of defects are parallel to the \([1\bar{1}0]\) as defined. A few defects were observed in the orthogonal direction. The defects exhibit a black and white contrast indicating that the dislocations are inclined with respect to the sample surface. As shown in Fig. 4, it was most common to observe two dislocations travelling from one buried QD in opposite directions through the sample thickness to the surface. Often these dislocations were terminated by a large relaxed island at the surface. For example in Fig. 4(b), the dislocation on the right is terminated at the surface by a large relaxed island but the dislocation on the left is not. This description is further supported by the occasional ‘v-shaped’ dislocation such as that identified by the arrow in Fig. 4(a). Also when the sample was tilted in a direction parallel to the \([110]\) (not shown) direction, all the dislocations became v-shaped. Cross-sectional TEM analysis will need to be performed to gain a further understanding of these dislocations as some of them appear dissociated. However they appear to be similar in character to those presented in Refs. [13,14]. The large island in Fig. 4(b) exhibits Moiré fringes, clearly confirming that it is relaxed. The spacing of these Moiré fringes and analysis of diffraction patterns indicate that the lattice spacing within the large island is \(1(\pm0.4)\%\) less than that of unstrained InAs. The large relaxed island is approximately 100 nm across its longest axis (10 times that of the smaller buried QDs) and has clearly defined crystallographic planes with a hexagonal shaped base consisting of sides parallel to the \([010]\), \([100]\) and only one of the two orthogonal \(\langle 110\rangle\) directions (the \([1\bar{1}0]\) in our convention). A second sample was grown without surface QDs and a similar dislocation density observed. Therefore, it is unlikely that the surface QDs are promoting dislocation formation but rather that the dislocations form first and are preferential nucleation sites for the surface QDs. As discussed in more detail in the next paragraph, these large surface islands are also observed by AFM.

Fig. 5 compares AFM images of uncapped QDs on the surface, grown using different buffer layers. It was found that these surface QDs significantly change morphology and coalesce as they are cooled down from the growth temperature and are not at all a true representation of the buried QDs. However it is interesting to examine the underlying surface. As the indium composition is increased from 0% to 7.5% ((a) to (b)) and then from 7.5% to 15% ((b) to (c)) the underlying surface becomes increasingly rough. The underlying buried QD layer is responsible for this surface roughness because below the 2D–3D transition, this surface roughness disappears (not shown). We suspect that the troughs in the AFM images are due to reduced growth rate near the dislocations discussed above. Large, faceted islands (major axis > 100 nm) similar to those observed by plan-view TEM are also observed in the AFM images. These are circled in white in Fig. 5(d) and commonly occur in pairs orientated along one of the \([110]\) directions. Therefore these may identify dislocation sites. They occur with a density approximately half that of the dislocations. However, not all the dislocations were terminated by two large islands and considering the AFM scan size this density is within reasonable agreement with that determined by TEM. These large islands typically occur within the troughs supporting the above argument. Similar effects have been observed in GaN materials [15] and high indium content InGaAs/GaAs quantum well systems [16].

4. Conclusions

In summary, we found that v-shaped dislocations with a density of \(\approx1.5(\pm1) \times 10^8 \text{ cm}^{-2}\) formed when InAs QDs were deposited onto a thin InGaAs buffer, whereas no such dislocations formed when the QDs were formed directly onto a GaAs buffer. These dislocations are correlated with both a reduction in photoluminescence intensity and a roughening of the GaAs capping layer. While it may be possible to avoid such dislocations by exploring further InGaAs buffer layer growth conditions, one should be cautious in using these InGaAs buffers when high QD optical efficiency is desired.
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References


Fig. 5. AFM images of the surface QDs deposited onto (a) GaAs, (b) a 7 ML In$_{0.075}$Ga$_{0.925}$As buffer layer and (c), (d) a 7 ML In$_{0.15}$Ga$_{0.85}$As buffer layer. Note that 300 nm below these surface dots is a buried QD layer. Please refer to Fig. 1. Pairs of aligned, large islands are circled in (d).
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