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Comparison of InAs quantum dots grown on GaInAsP and InP

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Abstract
We report on the growth of InAs quantum dots (QDs) on GaInAsP and InP buffers by metal–organic chemical vapour deposition on InP(100) substrates. Indium segregation and the As–P exchange reaction affect the QD nucleation and composition. The As–P exchange reaction has a more pronounced effect on the QDs grown on the InP buffer than on those grown on the GaInAsP buffer. A very thin (0.6 nm) GaAs interlayer grown between the buffer layer and the InAs QD layer consumes segregated indium and minimizes the As/P exchange reaction. Wavelength tuning from 1450 to 1750 nm covering the technologically important 1550 nm wavelength is also achieved for the InAs QDs grown with the thin GaAs interlayer.

(Some figures in this article are in colour only in the electronic version)

Self-assembled InAs quantum dots (QDs) on InP are very promising for long wavelength optoelectronic device applications (1.3–2 μm). Their potential use in optical fibre communications (1.3–1.55 μm) and atmospheric pollution control systems has attracted more efforts [1–5] to grow InAs QDs on the relatively small lattice mismatch substrate InP (3.2%). However the main difficulty associated with this material system is the As/P exchange reaction which occurs above 360 °C [6]. The efforts to grow InAs QDs on InP generally lead to InAsP alloy QDs having a large size inhomogeneity [7, 8] or quantum dashes or wires [9–11]. Even by only controlling the As or P overpressure, the formations of QDs and quantum wires on InP or an InAs layer due to the exchange reaction have been reported [10–13]. Earlier reports [14] suggest that the As/P exchange reaction stops after the top two monolayers (ML). However due to strain driven processes, the exchange reaction can affect the InAs QD growth [13] through the thin InAs wetting layer (WL) which is of ~2 ML [15]. The local variation in the strain field around InAs QDs and the presence of the asymmetric stress field might change the shape, size and composition of InAs QDs [12, 16]. The exchange reaction and indium segregation at the surface cause serious drawbacks like a large QD size fluctuation, alloying of QDs, a rough interface, a broad photoluminescence (PL) linewidth and low PL efficiency [4, 17–19]. For device applications, it is essential to control QD size distribution and PL wavelength by understanding and controlling the exchange reaction. In this study, we report the growth of InAs QDs by metal–organic chemical vapour deposition (MOCVD) on InP(100) substrates. The InAs QDs are grown on either an InP buffer or a lattice matched GaInAsP buffer layer on InP substrates. We show that the exchange reaction is associated with alloying which has a more pronounced effect on the QDs grown on the InP buffer than on those grown on the GaInAsP buffer. We also show that a very thin (0.6 nm) GaAs interlayer grown prior to the deposition of the InAs QDs can minimize the exchange reaction.

The InAs QDs are grown on (100) semi-insulating (SI) InP substrates using a horizontal flow MOCVD reactor at the pressure of 180 mbar. Trimethylindium (TMI), trimethylgallium (TMG), PH_3 and AsH_3 are used as precursors, and H_2 as a carrier gas. The InAs QDs are grown on InP or lattice matched $Ga_{0.25}In_{0.75}As_{0.54}P_{0.46}$ buffer layers at 520 °C using a V/III ratio of 15. The growth temperature for the rest of the structure is 650 °C. Following an oxide desorption step for the substrate at 700 °C for 10 min under PH_3, a 200 nm InP buffer layer is grown. Then InAs QDs are deposited directly on the InP buffer layer or on a 50 nm lattice matched GaInAsP ($\lambda_g = 1.28$ μm) buffer grown on top of the InP layer. In some samples, a 0.6 nm thick GaAs interlayer is grown at 650 °C before the growth of InAs QDs at 520 °C. The growth temperature is reduced from 650 to 520 °C under AsH_3 (PH_3 if the InP buffer is under the QD layer or AsH_3 and PH_3 if the GaInAsP buffer is under the QD layer) and the

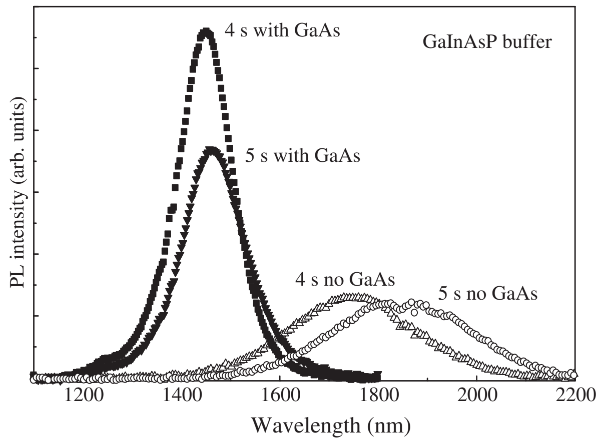


Figure 1. The room temperature PL spectra of the InAs QDs grown on the GaInAsP buffer with or without the 0.6 nm thick GaAs interlayer between the InAs QDs and the GaInAsP buffer.

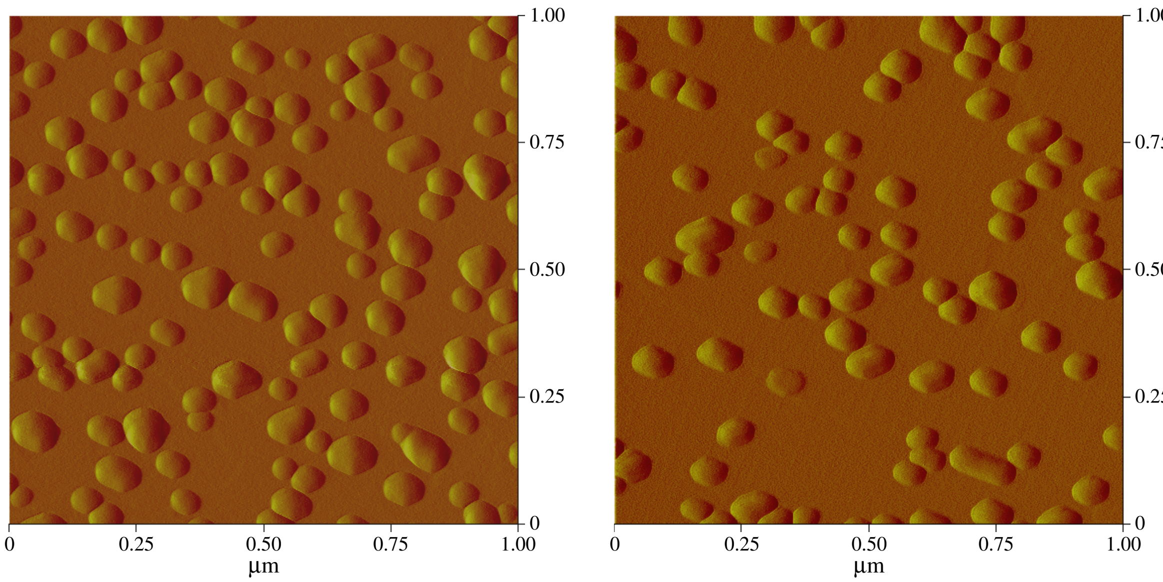
Table 1. Summary of the structures used in this study and the corresponding photoluminescence (PL) peak wavelengths of the InAs quantum dots (QDs).

Sample No	Buffer	Interlayer (GaAs)	QD deposition time (s)	PL peak wavelength (nm)
1	GaInAsP	—	4	1761
2		—	5	1867
3		0.6 nm	4	1451
4		0.6 nm	5	1461
5	InP	—	7	1431
6		—	8	1460
7		0.6 nm	7	1562
8		0.6 nm	8	1744

InAs QDs are grown for different times at a constant growth rate (0.16 nm s^{-1}). The InAs QDs are immediately capped without any growth interruption with a 200 nm InP layer while the temperature is increased to 650 °C. A summary of the QD structures and the corresponding photoluminescence (PL) peak wavelengths is shown in table 1. The same steps are followed to grow a top layer of InAs QDs for atomic force microscopy (AFM) studies. After the growth of the top InAs QD layer, the sample is cooled down to 400 °C under AsH_3. AFM measurements are performed with a multimode Nanoscope (Digital Instruments) in contact mode. Room temperature (RT) PL measurements are carried out by exciting with a 532 nm line of a frequency-doubled diode pumped solid state (DPSS) laser. The PL signal is collected by a thermoelectrically cooled InGaAs photodetector with a built-in preamplifier.

The room temperature (RT) PL spectra of the InAs QDs grown on the GaInAsP buffer layer are shown in figure 1. No QDs are formed on the GaInAsP buffer without any GaAs interlayer up to the InAs deposition time of 4 s. The AFM image of the top InAs layer and the plan-view transmission electron microscopy image of the buried InAs layer (not shown) confirm the presence of InAs QDs in the samples. The PL signal of these QDs is very broad with a peak at 1761 nm. The PL peak red-shifts to 1867 nm when the InAs QD deposition time is increased to 5 s, consistently with the increase in the QD size with more material deposition. However, with a 0.6 nm thick GaAs interlayer inserted between the GaInAsP buffer and InAs QDs deposited for 4 s, the PL peak blue-shifts to 1451 nm from 1761 nm and the PL intensity increases and linewidth decreases noticeably. The PL peak shift between 4 and 5 s samples is more in the case without a GaAs layer than for the ones with a GaAs interlayer due to the stabilization effect. In the latter case, the QD mean height does not increase much as the QD deposition time increases.

The AFM images of the InAs QDs grown for 5 s on the GaInAsP buffer with and without a GaAs interlayer are shown in figures 2(a) and (b), respectively. The average height of the QDs decreases from 10 nm to about 7.5 nm



(a) No GaAs

(b) With 0.6 nm GaAs

Figure 2. The AFM images of the InAs QDs grown for 5 s on the GaInAsP buffer. The thin GaAs interlayer thickness between the InAs QDs and the GaInAsP buffer underneath is either (a) 0 nm or (b) 0.6 nm. The scale is 1 μm × 1 μm.

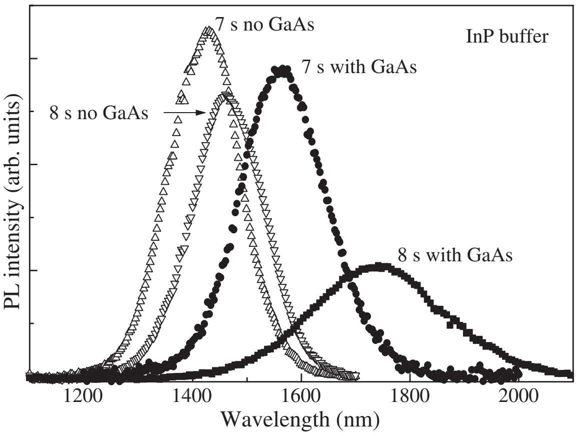
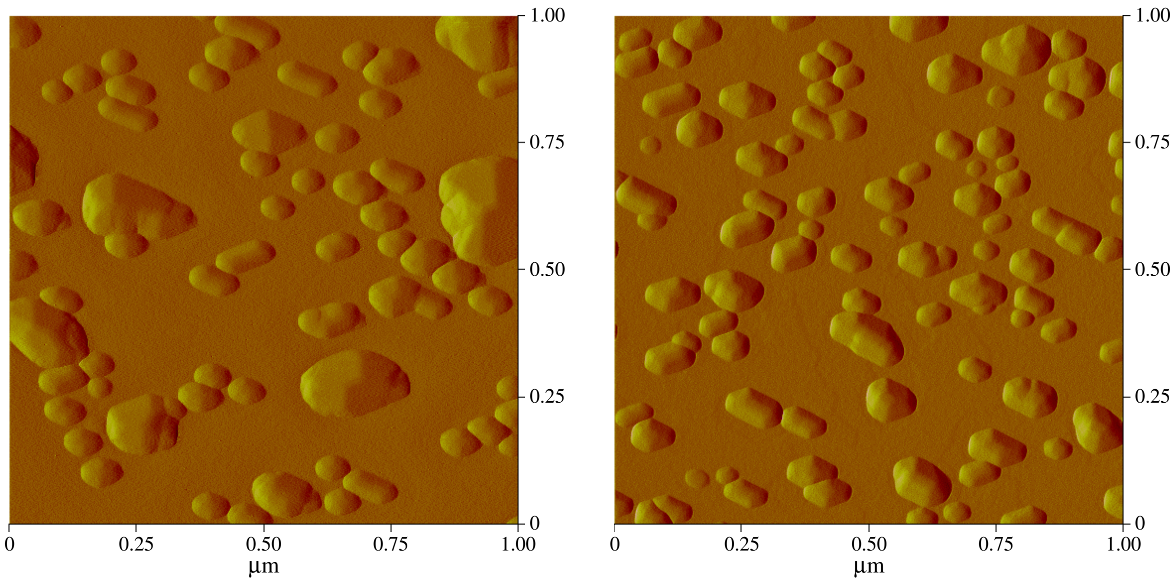


Figure 3. The room temperature PL spectra of the InAs QDs grown on the InP buffer with or without the 0.6 nm thick GaAs interlayer between the InAs QDs and the InP buffer.

when a thin 0.6 nm GaAs interlayer is inserted between the GaInAsP buffer and the InAs QDs layer. The mean diameter of the QDs remains almost same whereas the QD density decreases drastically from 1.1×10^{10} to 7.4×10^{9} cm^{-2}. Since the QD growth conditions are the same, the smaller QD height, diameter and density for samples grown with a GaAs interlayer indicates that the insertion of the thin GaAs layer results in less incorporation of the material. It is most likely that the increased surface energy due to the tensile strained GaAs layer (3.7% lattice mismatch on InP) reduces the incorporation efficiency of InAs. This effect is similar to those in some reports that showed that the incorporation of In during the deposition of an InGaAs layer reduces as strain increases [20]. The As/P exchange reaction is less likely since P atoms do not tend to replace As atoms in GaAs by breaking the more stable bond with Ga in GaInAsP. There could be some indium segregation on the GaInAsP surface. The segregated indium layer may provide more nucleation sites for InAs QDs formation, and thereby increase the QD density. During InAs growth, the segregated indium atoms may react with AsH_3 and form additional InAs which increases the QD size and its non-uniformity. The GaAs interlayer may consume this segregated indium layer blocking the additional source of InAs. As a result, besides the QD density and mean QD height, the QD size fluctuations also decrease causing much reduced PL linewidth after incorporation of the GaAs interlayer.

Figure 3 shows the room temperature (RT) PL spectra of the InAs QDs grown on the InP buffer layer. No QDs are seen under the AFM up to the InAs deposition time of 7 s. The InAs QDs grown for 7 s on the InP buffer layer without any GaAs interlayer show very good PL intensity centred around 1431 nm. The PL from the InAs QDs deposited for 8 s is red-shifted to 1460 nm. When a 0.6 nm thick GaAs interlayer is introduced between the InP buffer and the InAs QD layer, the PL from the InAs QDs grown for 7 s is red-shifted with a peak at 1562 nm and the PL intensity remains almost the same. The InAs QDs grown for 8 s on the GaAs interlayer also show a red-shifted, diminished and broad PL with a peak at 1744 nm.

The AFM images of the InAs QDs grown for 8 s on the InP buffer with and without a GaAs interlayer are shown in figures 4(a) and (b), respectively. The QDs grown directly on the InP layer are quite large with a mean height of 12 nm. The PL intensity of these QDs is quite good and the linewidth is quite narrow in contrast to their large size and broad size distribution. It is not clear whether the larger QDs are emitting and further studies are being carried out to find reasons for the narrow linewidth. When a 0.6 nm thick GaAs interlayer is inserted between the InP buffer and the InAs QD layer, the mean QD height decreases to 9.5 nm from 12 nm as shown in figure 4(b). The reduction in the QD mean height does not result in a blue-shift of the PL emission but a red-shift. Contrary to the GaInAsP buffer case, it is likely that InAsP



(a) No GaAs (b) With 0.6 nm GaAs

Figure 4. The AFM images of the InAs QDs grown for 8 s on the InP buffer. The thin GaAs interlayer thickness between the InAs QDs and the InP buffer underneath is either (a) 0 nm or (b) 0.6 nm. The scale is 1 μm × 1 μm.

alloy QDs are formed on the InP buffer. Due to the As–P exchange and the higher band gap energy of InAsP compared to that of InAs, the QDs directly grown on the InP layer are blue-shifted in comparison to the QDs grown on the GaInAsP layer. Furthermore, they are grown for longer times (more material deposition) in the case of InP buffer compared to GaInAsP buffer. But a thin 0.6 nm thick GaAs interlayer significantly reduces the As/P exchange reaction. Therefore InAs QDs form when a thin GaAs interlayer is grown on the InP buffer in comparison to the formation of InAsP QDs without the interlayer leading to a red-shift in PL emission peak wavelength. Compared to the InAs/GaInAsP QDs grown for 5 s on the GaAs interlayer as shown in figure 2(b), the InAs/InP QDs grown for 8 s on the GaAs interlayer have higher QD density (8.6×10^9 cm^{-2}) and QD mean height (9.5 nm) due to there being more material deposition.

The As/P exchange reaction occurs above 360 °C [6] and is expected to stop after 2 ML [14] of InAs layer deposition on the InP buffer but the strain associated with the InAs island formation enhances the exchange reaction [13, 16]. The InAs QD formation on InP substrates follows the strain driven Stranski–Krastanov growth mode. The exchange reaction can occur at the places through the thin InAs wetting layer which are not covered by the QDs. Due to their higher vapour pressure, P atoms desorb easily at these places particularly at the periphery of the QDs and free indium atoms [12, 16]. The free indium atoms migrate to the top of the dots driven by strain energy to decrease the total system energy and form excess InAs recombining with As atoms. This leads to taller QDs as noted in the AFM images. At the same time alloying can take place and it is likely that the composition of the QDs is not pure InAs but InAsP. In addition to this, alloying is more favourable since it relaxes part of the total strain energy [2, 8]. The alloying effect is more prominent in the InAs/InP QDs system than in the InAs/GaInAsP QDs system as observed in our case. In the $Ga_{0.25}In_{0.75}As_{0.54}P_{0.46}$ buffer, In–P bonds are only 35% of the total number of bonds; therefore the effect of InAsP alloying is less. When the GaAs interlayer is grown between the QDs and the GaInAsP buffer underneath, the height of the InAs QDs reduces and the dots become more uniform in terms of size and composition distribution due to suppression of the exchange reaction as noted in the AFM images and the narrower linewidth of the PL spectra. The reduction in the QD mean height results in a blue-shift of the PL emission wavelength, whereas in the InAs/InP QDs system, the insertion of GaAs causes a red-shift of the PL emission wavelength despite the smaller dot height. The QDs grown for 8 s on the InP buffer have broader QD size distribution and greater mean height (∼12 nm) than the QDs (mean height ∼10 nm) grown for 5 s on the GaInAsP buffer. Clearly all these effects show that As–P exchange leading to the formation of InAsP QDs is very prominent in the InAs QDs formed directly on the InP layer.

The lattice matched GaInAsP buffer and the InP buffer show very different characteristics as regards the InAs QDs formation. The InAs QDs start to form after 4 s of InAs deposition on the GaInAsP buffer whereas it takes 7 s for them to form on the InP buffer. The different amount of InAs material required to form QDs may be related to the different surface energy of the corresponding buffer. Indium segregation on the GaInAsP surface may help to form QDs on the GaInAsP buffer. In the InAs/InP QD system prominent alloying could change the compositions of both QDs and wetting layers to InAsP, which reduces the strain. As a result more material deposition is required to form QDs. Nevertheless the growth of a thin GaAs interlayer before the deposition of the InAs QD layer makes the InAs QDs behave in the same fashion and offers opportunities to tune the PL emission at RT.

In summary, the buffer layer composition affects the growth and composition of InAs QDs on InP substrates. The As–P exchange reaction is very prominent in the InAs/InP QD system. Indium segregation and the As/P exchange reaction affect the formation of QDs. A 0.6 nm thick GaAs layer grown between the buffer layer and the InAs QD layer consumes segregated indium and minimizes the As/P exchange reaction. It also offers the opportunity to tune the PL emission wavelengths over a wide range.

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