Growth, optical properties and device characterisation of InAs/GaAs quantum dot bilayers

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Abstract. The growth and optical properties of InAs/GaAs quantum dot (QD) bilayers are investigated, where the strain interactions between closely spaced QD layers are exploited to tailor the optical properties of the system. The underlying (seed) layer acts as a template for subsequent growth of the upper layer, whose properties can then be modified due to the greater freedom in the choice of growth conditions. Extension of the emission wavelength of the QDs is observed, to 1400 nm at room temperature for GaAs-capped bilayers and extending to 1515 nm for InGaAs-capped bilayers. The QDs in the second layer are highly uniform, resulting in an inhomogeneous broadening of \(<20\) meV and, for small separations between QD layers, efficient carrier tunnelling results in suppression of emission from the seed layer. Edge-emitting lasers incorporating QD bilayers operating either in the ground state or first excited state at 1340 nm at room temperature are demonstrated, showing comparable behaviour to QD lasers containing independent layers. Ground state lasing at 1425 nm at 250 K is also observed.

1. Introduction

During the last decade, In(Ga)As/GaAs quantum dots (QDs) have received considerable attention due to the predicted improvements in device performance that can be achieved because of their reduced dimensionality [1, 2] and novel behavior enabling development of new technologies such as single photon sources [3] and qubits for quantum information applications [4]. In addition, they provide extension of the operating wavelength of GaAs-based optoelectronic devices to 1300 nm and towards 1550 nm. Extension of the emission wavelength of InAs/GaAs QDs to the telecoms wavelengths of 1300 nm at room temperature has been achieved by a variety of techniques, including alternate layer epitaxy [5], use of low InAs growth rates [6] or, most commonly, by capping the QDs with a thin layer of InGaAs before subsequent GaAs growth [7] or encapsulating the QDs within an InGaAs quantum well (dots-in-a-well or DWELL systems) [8]. Edge-emitting InAs/GaAs QD laser diodes operating at 1300 nm were first demonstrated in 1998 [9] and are now reaching commercialization.

Further extension of the emission wavelength towards 1550 nm using the growth techniques mentioned above is problematic. For example, for growth at low InAs growth rates, there is a saturation in the emission wavelength of GaAs-capped QDs at around 1300 nm for the lowest growth rates [10, 11]. In order to extend the emission wavelength of InGaAs-capped QDs or DWELL systems beyond 1400 nm, very high In-content layers and/or a reduction in strain experienced by the QDs is
required [12]. There are only isolated reports of room temperature emission in excess of 1500 nm from InGaAs-capped QDs (grown by metalorganic chemical vapor deposition), using high In-content InGaAs caps [13, 14]. An alternative approach is to modify the cap by incorporating N or Sb [15, 16]. Although this further complicates the growth procedure, emission around 1550 nm has been achieved by these techniques. A further method that considerably reduces the strain on the QDs is to grow on InGaAs metamorphic buffers so that the surrounding matrix is InGaAs rather than GaAs. By doing this, room temperature lasing at 1515 nm has been demonstrated [17], although early devices have suffered from a high threshold current density (Jth) due to nonradiative carrier recombination at defects [18].

Rather than considering single QD layers acting independently, one can also manipulate the strain in the system by utilizing the strain effects of closely-stacked QD layers. In this paper we discuss the strain interactions between closely separated QD layers and their effect on the growth of subsequent layers, and a technique is presented for manipulating the growth of two nearby layers such that long wavelength emission can be achieved. In addition to the substantially extended emission wavelength, these QD bilayers have a number of novel properties that may be useful both for device applications and in providing an interesting physical system for further research. Edge-emitting lasers incorporating QD bilayers have been fabricated and initial characterization of these devices is compared with existing QD laser designs.

2. Growth of multiple layers of quantum dots

Because of the low areal coverage of QDs, in order to achieve sufficient gain required for many practical devices multiple layers of QDs must be incorporated into the active region. However growth of multiple layers such that the QDs in each layer are of comparable size, composition and strain state in order to achieve coincident emission is far from trivial. For small layer separations, underlying QDs provide a strain field that can penetrate through the spacer layer and influence the growth of upper layers. A smooth surface morphology and relatively large separations, typically of at least 40 nm, are required to ensure that the QD layers are independent and their optical properties are the same [19, 20]. This places a limit on the number of QD layers that can be incorporated into the active regions of many devices.

Many studies have demonstrated that when QD layers are closely stacked there are strong interactions between the QDs. Cross-sectional transmission electron microscopy (TEM) and scanning tunneling microscopy (STM) show that the QDs in upper layers are vertically correlated with underlying QDs, and also that there is an increase in QD size in upper layers [21, 22]. This can also be observed by in situ reflection high-energy electron diffraction (RHEED) during growth. This is illustrated in figure 1, which shows RHEED measurements of the InAs coverage required to reach the 2D – 3D transition for QD formation ($\theta_{\text{crit}}$), obtained during the growth by molecular beam epitaxy of two closely spaced QD layers. The QD layers were each formed by deposition of 2.4 ML InAs onto an annealed GaAs surface at a growth rate of 0.014 MLs$^{-1}$. Following growth of the first layer, the QDs were capped by a GaAs spacer layer and then this surface was annealed for 10 minutes at 580 °C under an As flux to desorb segregated In and smooth the surface before growth of the second layer, as confirmed by the clear GaAs surface reconstruction observed by RHEED. Figure 1(a) shows $\theta_{\text{crit}}$ for the second layer as a function of GaAs spacer layer thickness, for QD layers grown at 510 °C. As the separation between the QD layers is reduced, a reduction in $\theta_{\text{crit}}$ is observed, reaching a minimum for a spacer layer thickness of 7 nm. This effect cannot be attributed to segregated In on the surface before deposition of the second layer as it is observed despite the annealing of the surface before growth of the second layer. The increase in $\theta_{\text{crit}}$ that is observed for even smaller separation is due to incomplete capping of the first layer and subsequent In loss during the annealing process [23].

For two QD layers separated by 10 nm GaAs, $\theta_{\text{crit}}$ is reached ~25% quicker for the second layer compared to the first and previous TEM studies of stacked QD layers with similar separation show the degree of vertical correlation of the QDs is close to unity [21]. The influence of this templating effect of the lower layer despite changes in the growth conditions is shown in figure 1(b). For the first layer,
θ_{crit} increases with temperature, as is usually observed for growth of single QD layers. However for the second layer, separated by 10 nm from the first, θ_{crit} remains approximately constant over the temperature range considered. This indicates that preferential nucleation above buried QDs dominates over thermal effects in the process of QD formation.

The vertical correlation of QDs in closely stacked layers is commonly attributed to strain fields due to buried QDs penetrating the spacer layer and causing a modulation of the local strain at the surface during growth of subsequent layers. Regions of tensile strain are located above buried QDs, which create preferential sites for QD nucleation: since InAs growth is more likely in the regions immediately above buried QDs, 3D island formation will occur with deposition of less material than is required for the case of uniform coverage across the surface, leading to the earlier observation of the 2D – 3D transition and resulting in larger QDs than if the same amount of material is deposited.

Despite the increase in size of QDs in the upper layer, photoluminescence (PL) studies have shown a blueshift in the emission from upper layers compared to single (lower) layers for QDs grown under the same conditions of temperature and growth rate [24, 25]. This is attributed to increased intermixing induced by strain gradients between regions of tensile strain above buried QDs and regions of compressive strain in between buried QDs [26]. Increased intermixing in upper layers of stacked QD systems has been observed using TEM or STM for both Ge/Si [27] and InAs/GaAs QDs [28]. However, as demonstrated in the next section, the template effect of the lower QD layers shown in figure 1(b) allows greater freedom in the choice of growth conditions in the upper layer and this can be used to compensate or suppress the increased intermixing.

### 3. InAs/GaAs QD bilayers

In order to circumvent the blueshift of emission in closely-stacked QD layers, various techniques have been employed to achieve either coincident emission from the layers or to further redshift the emission of the upper layer. Due to the high degree of vertical correlation of QDs in closely stacked layers, the first (seed) layer acts as a very efficient template for QD nucleation in the second (upper) layer and growth conditions for the second layer can be modified without affecting the QD density. The strain-induced intermixing can then be counteracted by increasing the InAs coverage [28, 29], the growth rate [30] or, crucially, the growth temperature [31]. A reduction in growth temperature will suppress In/Ga intermixing during QD formation and during capping so the QD composition will be close to pure InAs. Figure 2 shows 10 K and 300 K PL spectra obtained from a QD bilayer grown using a reduced growth temperature and increased InAs coverage in the second layer. The bilayer was grown...
by initial deposition on an annealed GaAs surface of a seed layer of 2.4 ML InAs grown at a growth rate of 0.014 MLs\(^{-1}\) at a temperature of 480 °C, giving a QD density of 3 \(\times 10^{10}\) cm\(^{-2}\), which was then capped by a GaAs spacer layer of 10 nm, also grown at 480 °C. The surface was then annealed under an As flux at 580 °C for 10 minutes prior to deposition of the second layer of 3.3 ML InAs at the same growth rate but at a reduced temperature of 467 °C. The QDs were capped by 15 nm GaAs at 467 °C before the temperature was ramped to 580 °C for growth of a further 85 nm GaAs. A similar sample containing only the seed layer was also grown and figure 2 also shows PL spectra obtained from this sample for comparison.

![Figure 2. (a) 10 K, (b) 300 K PL spectra obtained using HeNe laser excitation from a GaAs-capped QD bilayer and a single QD layer grown under the same conditions as the seed layer](image)

Due to the low growth rate used for these samples, the ground state PL emission of the single (seed) layer sample is at a relatively long wavelength (1160 nm at 10 K, extending to \(\sim\)1250 nm at room temperature) with a narrow inhomogeneous broadening of 25 meV at 10 K, indicating that the QDs are fairly large and In-rich and the size distribution of the QDs is highly uniform. However emission from the bilayer sample is further redshifted by almost 100 nm compared to the seed layer alone and the inhomogeneous broadening is reduced to only 14 meV at 10 K, rising to 17 meV at room temperature. The extremely narrow emission linewidth observed for these structures is especially attractive for device applications [32]. STM analysis of uncapped QDs in the upper layer shows a significant reduction in the fluctuation in QD height when the temperature is reduced for the growth of the second layer [31], and suppression of In/Ga intermixing during capping at low temperature will also minimize variation in QD composition.

Suppression of emission from the seed layer is also observed in the bilayer PL spectra, due to efficient carrier transfer from the seed layer to the upper layer QDs. Note that the QD layers are located 100 nm from the sample surface and so significant photo-excitation of carriers into the GaAs matrix both above and below the QDs is expected, allowing carrier capture into QDs in both layers (the penetration depth of a HeNe laser into bulk GaAs is 220 nm at 5 K [33]). Previous time-resolved PL studies of closely stacked InAs/GaAs QDs show that tunneling times between vertically correlated QDs can be rapid, depending on the spacer layer thickness (as short as 20 ps) [34]. Carrier relaxation times within an InAs/GaAs QD are also rapid (~ps) [35, 36] and characteristic times for both mechanisms are much shorter than the radiative lifetime for excitons in QDs (typically ~ns [37]), so
carriers are likely to tunnel from the higher energy ground states of the seed layer QDs into excited states of the upper layer QDs, relax to the ground state and undergo radiative recombination from these QDs, thus suppressing seed layer emission and also acting as an additional carrier capture pathway for the second layer QDs. As the separation between layers in the bilayer structure is increased, electronic coupling between the QD layers is inhibited but strain coupling of the QDs leading to preferential nucleation sites during growth of the upper layer may still persist [38].

Room temperature emission from the InAs/GaAs bilayer shown in figure 2 is well in excess of 1300 nm, despite the QDs being surrounded by a GaAs matrix. By careful choice of growth conditions for both the seed and upper layers of the bilayer, room temperature emission approaching 1400 nm can be achieved [31], well beyond the emission wavelength of single low-growth-rate QD layers and without incorporation of InGaAs strain reducing layers. However InGaAs capping of the upper layer QDs can still be used to further extend the emission wavelength of the bilayers, as detailed in the next section.

4. Extension of bilayer emission to >1500 nm

As discussed above, the seed layer acts as a very effective template to set the density of QDs grown in the upper layers of the bilayers, and so a variation of the growth conditions of the seed layer should lead to corresponding changes in the properties of the upper layer. For example by changing the growth temperature of the seed layer, the QD density can be varied by an order of magnitude. Because of the high degree of vertical correlation in the bilayers, this will lead to a comparable change in the QD density in the upper layer. This is shown in figure 3, which presents results of atomic force microscopy (AFM) analysis of uncapped seed layers or second layers of bilayers grown following the procedure outlined in section 3 above but changing the growth temperature of the seed layer.

![Figure 3](image-url)

**Figure 3.** Variation of the QD density in the seed layer (diamonds) and the second layer (triangles) of InAs/GaAs QD bilayers with change in the seed layer growth temperature. Note that the growth temperature of the second layer remains the same (467 °C) for all bilayer samples.

As the growth temperature of the seed layer is varied from 480 to 515 °C, a reduction in the QD density in both the seed layer and the second layer is observed from $3 \times 10^{10}$ cm$^{-2}$ to $4 \times 10^9$ cm$^{-2}$. There is also a concomitant increase in QD height (not shown) in both layers, from an average seed
layer QD height of 6 nm at 480 °C to 15 nm at 515 °C, because the same amount of deposited In is distributed amongst fewer QDs and also (for the seed layer) possibly due to increased Ga inclusion from the substrate at higher temperatures [39]. As the QD size in the second layer is increased, one would expect the emission wavelength to be extended (the second layer is grown at 467 °C so compositional variations due to intermixing should be minimized), and this indeed is observed, as shown in the inset to figure 4. For GaAs-capped bilayers, a variation in peak room temperature emission from 1340 to 1400 nm is seen as the seed layer growth temperature is changed.

![Figure 4](image)

**Figure 4.** Room temperature PL spectrum obtained from an InAs/GaAs QD bilayer with seed layer grown at 505 °C and the second layer capped by 4 nm In$_{0.26}$Ga$_{0.74}$As. Inset: variation of peak room temperature emission from InAs/GaAs QD bilayers capped either by GaAs (diamonds) or additionally by an InGaAs strain-reducing layer (triangles) with change in seed layer growth temperature.

As mentioned above, incorporation of an InGaAs capping layer into the bilayer structure further extends the emission wavelength of the QDs, by approximately 100 nm, which for the bilayers grown with a higher seed layer temperature can then exceed 1500 nm. Figure 4 shows a room temperature PL spectrum obtained from a bilayer grown following the procedure outlined earlier but with a seed layer grown at 505 °C and for which the upper QD layer was initially capped by 4 nm In$_{0.26}$Ga$_{0.74}$As before subsequent GaAs growth. A narrow PL linewidth of 22 meV at room temperature is observed, as well as suppression of the seed layer emission, which persists to low temperatures (not shown).

These long wavelength QD bilayers have not yet been included in device structures, but previous designs with ground state emission at 1340 nm and 1450 nm at room temperature have been incorporated into edge-emitting lasers, as described in the next section.
5. Bilayer lasers

Edge-emitting lasers incorporating GaAs-capped and InGaAs-capped QD bilayers have been fabricated. The devices consist of a conventional p-i-n structure with an undoped GaAs active region of 500 nm containing three bilayers each separated by 50 nm GaAs, surrounded by 1500 nm doped AlGaAs cladding layers. The wafers were processed into 5mm long ridge waveguide lasers with a shallow-etched ridge width of 5 µm. The inset to figure 5 shows cw electroluminescence (EL) spectra as a function of bias current obtained at a temperature of 10 °C from the GaAs-capped bilayer laser, which contains three bilayers grown under the same conditions as described in section 3. A low ground state lasing threshold current density $J_{th} = 70 \, \text{Acm}^{-2}$ is measured [40]. As the current is further increased, lasing from the first excited state is also observed. Such behaviour has been observed for other types of QD laser [41] and is not a consequence of the bilayer design, and has been attributed to either a carrier relaxation bottleneck within the QDs [41, 42] or to competition between transverse modes in the laser cavity [40].

![Figure 5. Variation of threshold current density with temperature for three QD lasers operating around 1300 nm at room temperature: a laser containing 3 bilayers (diamonds), a laser containing 5 independent QD layers (squares) and a laser containing 5 DWELL layers. Inset: 10 °C cw EL spectra as a function of bias current obtained from the bilayer laser.](image)

Figure 5 also shows the variation of $J_{th}$ with temperature for the bilayer laser and for two other lasers: a laser grown to a similar design but with an active region containing 5 single QD layers (grown under similar conditions as the seed layers of the bilayers), separated by 50 nm and a laser containing 5 DWELL layers, also separated by 50 nm. All three lasers show similar behavior with respect to temperature, with the laser threshold current remaining stable through low temperatures...
(corresponding to a high characteristic temperature $T_0$ [1]) but increasing rapidly above $\sim 260$ K (low $T_0$ around room temperature). This is commonly observed for QD lasers [43] and $T_0$ near room temperature can be improved by introducing modulation p-doped layers near the QDs [44].

Figure 6. 300 K EL spectra obtained from the QD laser containing independent layers, the GaAs-capped QD bilayer laser and an InGaAs-capped QD bilayer laser (pulsed mode, 2 µs pulses, 0.2% duty cycle). Inset: Ground state lasing from the InGaAs-capped bilayer laser at 250 K.

The laser containing the InGaAs-capped bilayers was fabricated to the same design as the GaAs-capped bilayer laser described above. Each bilayer was grown using a seed layer growth temperature of 492 °C, with the upper layer initially capped by 4 nm In$_{0.26}$Ga$_{0.74}$As before subsequent GaAs growth. Figure 6 shows lasing spectra obtained using a pulsed current source from this device, with spectra from the GaAs-capped bilayer laser and the seed layer laser for comparison. The InGaAs-capped bilayer laser had a considerably greater $J_0$ than the other devices and only operated in the excited state at room temperature. It is currently unclear whether this was due to the InGaAs capping or due to contamination of the MBE machine during growth. However the excited state lasing was at a wavelength of almost 1350 nm: this offers the exciting possibility of fabricating devices for operation in the 1300 nm region using the QD excited states, which may provide advantages in increased gain (due to the increased degeneracy of the excited state), output power and modulation speed. When the device was cooled to 250 K, ground state lasing was achieved at a wavelength of 1425 nm, which is comparable to the emission wavelength of QD bilayer lasers reported previously [45].

6. Conclusion
By exploiting the growth conditions that occur when two QD layers are closely stacked, it is possible to manipulate the optical properties of the QDs and achieve long wavelength emission within a GaAs matrix. These bilayer systems also exhibit other novel properties, including an exceptionally narrow inhomogeneous broadening of $<20$ meV at room temperature and efficient carrier transfer between layers. They offer considerable benefits for device applications but are also an interesting system for investigating strain aspects of semiconductor nanostructures.
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