Defect dissolution in strain-compensated stacked InAs/GaAs quantum dots grown by metalorganic chemical vapor deposition

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We report a highly effective growth technique to both dissolve large islands and prevent further defect propagation in closely spaced (15 nm) stacked quantum dot (QD) active regions while maintaining an emission wavelength >1.3 µm. Island dissolution is accomplished via an In flush, which is an AsH₃ pause inserted into the growth sequence just after each QD layer is capped. The low V/III ratio enables the flushing of surface In atoms from the defect sites while the fully capped QDs remain intact. This technique eliminates the need for in situ annealing that activates the In flush in other growth scenarios and results in large emission blue shift. Strain propagation within the closely spaced QD stacks is reduced by GaP strain-compensation layers. Room-temperature photoluminescence confirms ground-state emission wavelength >1.34 µm. Atomic force microscopy and transmission electron microscopy confirm improved surface morphology and crystalline quality of stacked QD active regions. The resulting structures are suitable for long-wavelength lasers, especially vertical cavity surface-emitting laser applications in which high modal gain is attractive. © 2005 American Institute of Physics. [DOI: 10.1063/1.2042638]

Self-assembled InAs/GaAs quantum dots (QDs) have attracted much attention in modern semiconductor laser application due to the unique properties of zero-dimensional systems, low threshold current density and temperature sensitivity. However, many device applications require higher gain than a single QD layer can provide, thus stacked QD layers become attractive. Stacking the QD layers within the active region grown by molecular-beam epitaxy increases ground-state modal gain near 1.3 µm, resulting in ground-state lasing and larger Tₚ. In most demonstrated structures, the QD layers are typically separated by >30 nm. This configuration works well for some edge emitting applications, but for other applications, such as high bandwidth lasers and vertical cavity surface-emitting lasers (VCSELs), a high modal gain is attractive and the QD layer separation should be minimized.

In comparison, QDs for 1.3 µm wavelength emission grown using metalorganic chemical vapor deposition (MOCVD) have been more difficult due to the high density of polycrystalline coalesced islands that form with increased strain and complicated surface environment. Both Kaian der et al. and Tatebayashi et al. have employed an In-flushing method that utilizes an annealing step to dissolve the large coalesced islands. In both cases, the anneal leads to ground-state lasers emitting at λ>1.24 µm. Other groups have also reported MOCVD-grown stacked QD lasers near 1.3 µm. Kim et al. has achieved lasing at λ=1.28 µm by using InAs/InGaQD cladding InGaP lasers. Our group has previously demonstrated a thin GaP (Δd₀=3.8%) tensile layer embedded in a GaAs matrix to partially compensate the compressive strain of the InAs (Δd₀=7%) QD layer result in ground-state lasing at λ=1.25 µm.

In the previously reported In-flushing method, island dissolution is accomplished by increasing the wafer temperature just after the InAs QDs are capped with a thin GaAs barrier. During this anneal, the capped InAs QDs are completely covered by the GaAs barrier and remain intact, while the large InAs defect clusters are exposed to the surface and will be partially evaporated. The disadvantage of this method is the In/Ga intermixing driven by the increased temperature that results in large blueshift and degraded emission efficiency.

In this letter, we describe an In flush that employs an AsH₃ pause to dissolve the large defective islands rather than an anneal. Our QD stacks also include GaP strain-compensated (SC) layers to reduce defect propagation in the stacked QD ensemble and allow a total interlayer spacing of only 15 nm. Our samples are grown by MOCVD at 60 Torr using trimethylgallium, trimethylindium, tertiarybutylphosphine, and arsine (AsH₃). Growth is initiated on a GaAs (001) substrate with a 300 nm GaAs layer at 680 °C, then the temperature is reduced and stabilized for active region growth within the range of 450–520 °C. The growth procedure for the active region is described in Figs. 1(a)–1(e). Each QD layer consists of a 5 monolayer (ML) In₀.₁₅Ga₀.₈₅As buffer followed by a 3 ML InAs QD layer, as illustrated in Fig. 1(a). A postnucleation AsH₃ pause is used after the growth of each QD layer to reduce defect density and improve QD uniformity. The first QD layer is capped with a 7 nm In₀.₁₅Ga₀.₈₅As and 4 nm GaAs layer shown in Fig. 1(b). Then, the In-flushing step (AsH₃ pause) is initiated, Fig. 1(c), at the same growth temperature. During this step, all sources are switch off, including AsH₃, allowing the In atoms from large uncapped defect cluster to diffuse along the GaAs surface and evaporate from the structure. The pause time for this step is varied from 60 to 150 s. Growth is resumed with a 4 ML GaP SC layer and another 4 nm GaAs barrier followed by the next InAs QD layer shown in Figs. 1(d) and 1(e). The remaining In atoms on the GaAs surface are trapped in the GaP SC layer. For this study, a total of five QD stacks are grown in this manner and finally capped with 80 nm of GaAs.
The effect of In flushing on the optical properties of five-stack QD samples has been studied as a function of In-flushing times including 60, 90, 120, and 150 s. The room-temperature photoluminescence (RTPL) is characterized using a 5 mW HeNe pump laser (20 mW/cm²), a lock-in amplifier, and an InGaAs detector. The important trends are noted in Fig. 2. The ground-state emission wavelengths are observed between λ = 1315 nm and λ = 1335 nm with a slight variation in full width at half maximum from sample to sample within the range from 50 to 65 meV. The photoluminescence (PL) intensity increases with In-flushing time from 60 to 90 s, then decreases for 120 s and 150 s. The increase in intensity with the flushing step is attributed to the reduction of defect clusters, which reduces the number of nonradiative recombination centers in the structure. The decrease in PL intensity associated with a longer flushing time is most likely caused by As vacancies at the growth surface and dissolution of the QD material indicated by the small blueshift of PL wavelength associated with the longer pause times.

Figure 3 shows cross-sectional transmission electron microscopy (TEM) images of a five-stack QD active region with 90 s flushing steps and 4 ML GaP SC layers. In this structure, the QDs are vertically aligned to form a column by the residual strain field from lower QD layers. This strain accumulation can initiate large defect clusters in a regular stacked structure. The flushing step serves to dissolve the large polycrystalline islands and reduce defect formation and defect propagation. During the flushing step, In atoms on top of the large island are removed as shown by the brighter area atop the dissolved island shown in the inset of Fig. 3. In fact, the In atoms appear to leave the island center resulting in a flattened and slightly voidlike structure. These flattened islands obtained in our structure are similar to the quantum-ring structures already documented through experimental and theoretical reports. Further optimization of the In flush will reduce the occurrence of voidlike structures in laser active regions.

Figure 4 shows root-mean-square (rms) roughness of the five-stack QD structure as a function of flushing time obtained from (1 μm x 1 μm) atomic force microscope (AFM) images. The flushing steps result in a significantly smoother surface as the crystalline defect density is reduced. The rms roughness decreases linearly from 1.251 nm to 0.114 nm as the flush time is increased from 0 s to 150 s and corroborates a reduced defect density in the structure. The insets of Fig. 4 show the three-dimensional surface morphology of the three structures with 150 s flushing (rms roughness = 0.114 nm), 90 s flushing (rms roughness = 0.626 nm), and without (rms roughness = 1.251 nm). Without In flushing, an undulating surface is visible, and with In flushing the surface is atomically smooth. The improved surface morphology is
expected to significantly reduce optical loss in the laser structure.

Finally, we characterize the formation of surface QDs grown atop a five-stack active region both with and without the In flush. Figures 5(a)–5(d) show AFM images (2 μm × 2 μm) of surface QDs without SC layers, with SC layers, and a 90 s In-flush, and single QD layer, respectively. The five-stack sample [Fig. 5(a)] without SC has a high defect density of 2.1 × 10⁹ cm⁻² and surface undulations with QD density ~2.4 × 10¹⁰ QDs/cm². The defect density with SC, shown in Fig. 5(b), remains in the 10⁸ cm⁻² regime which is comparable to single stack QD shown in Fig. 5(d). The addition of In flushing along with SC layer, as in Fig. 5(c), results in significantly higher material quality and a QD ensemble free of coalesced islands. From comparing QD size in Figs. 5(b) and 5(c), we see that the average QD size is not strongly affected by the In flush and remains stable in width (~35 nm) and height (~7.5 nm) with QD density ~5 × 10¹⁰ QDs/cm² for both structures. The average QD size increases by ~10% in width and ~35% in height compared to single QD layer.

In conclusion, we have reported a technique for defect reduction in closely stacked QD active regions by introducing an In-flushing step in combination with GaP SC layers. These two steps each have a unique purpose in the structure. The GaP SC layers reduce residual strain propagation allowing a dense QD stack and the In flush reduces the density and propagation of defective islands that are common in MOCVD growth. The In flush described in this work effectively dissolves the defective islands via an AsH₃ pause rather than an anneal step that also shortens the emission wavelength. The RTPL shows that the resulting emission wavelength using this method remains >1.34 μm. Both improved morphology and defect-free structures suggest that this growth technique is suitable for long-wavelength emission laser applications. Furthermore, the QD layer separation of only 15 nm will produce an attractive active region for VCSEL applications.