Growth of low density InP/GaInP quantum dots

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Abstract. We demonstrate low density InP quantum dot growth by metalorganic vapour phase epitaxy on (100) GaAs substrates with a 3° misorientation towards ⟨111⟩. Using microphotoluminescence characterisation, we identify the transitional region from single to bimodal quantum dot size distributions where InP deposition thicknesses, over a range of ≈ 1.1˚A, provide a region of spatially and spectrally isolated quantum dots suitable for single dot studies.

Strong confinement effects and the resulting isolation of charge-carriers from the surrounding bulk material in individual quantum dots (QDs) have, in the last 10 years, received close attention[1]. The resulting phenomena which have been revealed, show the suitability of both InGaAs/GaAs and InP/GaInP QDs to a wide variety of interesting applications, ranging from single photon emitters [2, 3, 4] to spin qubits for quantum information processing [5, 6, 7, 8]. Recent progress in optically pumped InP/GaInP QDs, grown by metalorganic vapour phase epitaxy (MOVPE), has recorded high degrees of nuclear spin polarisation ≈ 65% as well as ultra-long nuclear depolarisation times up to 5000s[9, 10, 11]. However, multi-modal distributions of QD sizes are common - with QD heights ranging from 1 to 15nm[12]. This leads to spatially non-isolated QDs and therefore additional relaxation pathways and uncontrollable interactions with highly charged neighbours[13]. Thus growth of InP/GaInP QDs with a single distribution of QD sizes is essential for single dot studies.

In this work, we demonstrate crystal growth leading to reproducible and finely controlled samples containing only small QDs with densities lower than 10⁹cm⁻², avoiding multi-modal QD size distributions. We discuss the growth requirements for such structures and the influence of uni-directional reactant flow on the QD growth. We identify InP deposition thicknesses over a range of ≈ 1.1Å, where the transition from single to a bimodal QD size distribution provides a region of spectrally low density QDs.

Growth was performed using low-pressure MOVPE in a horizontal flow quartz reactor, with two wafers exposed side-by-side (upstream and downstream) for each growth run. All samples were grown on (100) GaAs substrates with a 3° misorientation towards ⟨111⟩ and an InP growth rate of 1.1Å/s. The growth temperature of the GaAs buffer and bottom GaInP layer was 700°C. Before deposition of the InP (QD) and GaInP (capping) layers, the wafer was cooled to 650°C. For samples grown with InP deposition time (tdep) of 10s, the temperature for the GaInP capping layer was 700°C. The grown GaInP layers were nominally lattice matched to GaAs.
Figure 1. Low temperature \(\mu PL\) spectra of InP/GaInP QDs \((P = 15\mu W)\) at two positions are shown. Comparing the spectra at an upstream (black) and downstream (grey) position, the change in size distribution of the QDs depending on position in the growth chamber is observed. Inset at \(P = 1\mu W\).

All samples were measured at a temperature of 15K using a micro-photoluminescence (\(\mu PL\)) set-up. A laser with a wavelength of 543nm or 633nm, focussed onto a 2\(\mu m\)-diameter spot on the sample, was used to excite carriers in all spectra presented. \(\mu PL\) signal was measured with a high spectral resolution using a double spectrometer/CCD detection, enabling detection of photoluminescence (PL) from individual QDs.

Figure 1 shows typical \(\mu PL\) spectra recorded at relatively high excitation power, \(P = 15\mu W\), for previously studied QD samples grown with \(t_{dep} = 10s\). As seen for both upstream (black line) and downstream (grey line) samples, a pronounced bimodal size distribution is observed with two broad peaks centred at 1.77 and 1.65eV, corresponding to small and large QDs, respectively. Sharp peaks corresponding to PL of individual QDs are observed in the range 1.71–1.77eV at a lower excitation power, \(P = 1\mu W\), for which population of excited states in QDs is suppressed [see Figure 1 inset]. However, even at such low power the peak at 1.65eV remains broad and featureless implying markedly higher densities of large QDs. Figure 1 also shows that the relative intensity of the low energy peak decreases for the downstream sample - indicating a lower density of large QDs forming at this point in the growth chamber.

The effect of reducing \(t_{dep}\) is shown in Figure 2, measured at \(P = 0.15\mu W\). At the shortest growth time, 2.0s (grey line), we measure emission from only small QDs with a narrow size distribution peaked at 1.85eV and the wetting layer is observable at 1.89eV. As \(t_{dep}\) is increased the QD PL shifts to lower energy, becomes spectrally broader and wetting layer emission is no longer observed. At \(t_{dep} = 3s\) (upper grey line), a weak peak at 1.64eV is first observed, corresponding to initial formation of large QDs.

The optimum conditions for the growth of low density QDs with a single size distribution, are reached at \(t_{dep} = 3s\). Between \(t_{dep} = 4\) and 2.5s, over a range of InP deposition times, \(\Delta t_{dep} \approx 1s\), we observe a relatively small number of individual QD peaks, distributed in a wide spectral range, allowing clear observation of optical properties of individual QDs. This corresponds to a change in the InP deposition thickness of \(\approx 1.1\AA\) and the QD density changes from \(1.1x10^9\) to \(0.8x10^9\) cm\(^{-2}\), for \(t_{dep} = 2.5\) and 3s respectively (where the density is estimated by counting the number of lines in the spectra at low powers).

We note that the observed range of deposition times which leads to growth of suitable samples, \(\Delta t_{dep} \approx 1s\), is large in comparison to the mechanical growth-control time. This results in very reproducible growth confirmed in our further growth experiments. We also note a smooth variation of QD density over the length of the reaction chamber is observed. As the measurement position is moved further downstream, the QD and bulk GaInP PL peaks shift to higher energy. This may be due to reduced concentration of In and Ga reactants on passing down the growth chamber.
In conclusion, MOVPE growth conditions for spectrally and spatially isolated InP/GaInP QDs have been achieved. The obstacle of multi-modal size growth has been overcome and densities of less than $10^9 \text{cm}^{-2}$ have been observed. We find a reproducible, smooth transition in QD size distribution and density for varying InP deposition times and position along the growth chamber. A wide range of InP deposition times corresponding to $\approx 1.1 \text{Å}$ change in deposition thickness, where conditions are suitable for single dot studies, has been found.

Acknowledgments
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