Optimization of low density InP/GaInP quantum dots for single-dot studies

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Abstract. We achieve well-controlled and reproducible growth by low-pressure metalorganic vapor phase epitaxy (MOVPE) of low density InP/GaInP quantum dots optimized for single-dot physics and applications. We overcome the common occurrence of multi-modal distributions of quantum dot sizes by optimizing the growth on (100) GaAs substrates with a 3° misorientation towards ⟨111⟩. In contrast to other epitaxial techniques for quantum dot growth, very controllable dependence of the quantum dot sizes and densities on the nominal thickness of the InP layer is observed, enabling highly reproducible growth.

1. Introduction

In the last 10 years, experiments on individual quantum dots (QDs) have revealed a wealth of effects due to strong confinement and the resulting isolation of charge-carriers in these nano-structures from the surrounding bulk material [1]. Of particular interest are self-assembled QDs, including InGaAs/GaAs and InP/GaInP QDs, which have been widely researched for applications ranging from single photon emitters [1, 2, 3, 4, 5] to spin qubits for quantum information processing [1, 6, 7, 8]. QDs in both material systems have recently revealed novel spin phenomena of current high interest in semiconductor research and exhibit properties important for spin control on the nano-scale in solid state quantum devices [1, 6]. In optically pumped InP/GaInP QDs, grown by metalorganic vapor phase epitaxy (MOVPE), record high degrees of nuclear spin polarization ≈ 65% as well as ultra-long nuclear depolarization times up to 5000s have been observed[9, 10, 11].

However, samples used in these experiments commonly contain multi-modal distributions of QD sizes, consisting of short wavelength (660 – 730nm), small QDs and longer wavelength (730 – 770nm), large QDs [11, 5, 12, 13, 14, 15]. Although these reproducibly grown samples allow access to individual QDs in the short wavelength range, their properties are influenced uncontrollably by interactions with high density, large QDs. These large QDs have been shown to accumulate high numbers of charges at low temperatures[13], leading to charge instability and additional spin relaxation pathways in the neighboring small QDs. This makes the growth of single-modal distributions essential for single dot studies. In addition, the ability to reproducibly grow interacting double InP/GaInP QDs is enhanced by reproducible single-mode growth, thus
avoiding the wide height variation of multi-modal growth, where heights from 1 to 15nm have been observed[5, 12, 13, 14, 15].

In this letter, we report on a MOVPE crystal growth optimization study which enabled us to avoid formation of high densities of large QDs, leading to optimized samples containing only small QDs with densities lower than $10^{9}\text{cm}^{-2}$. We investigate the growth conditions required to reproducibly and accurately control both the size and the spatial distribution of single InP/GaInP QDs. This is shown to be achievable by selecting misoriented GaAs substrates and an appropriate InP deposition thickness ($d_{\text{InP}}$). We show that on (100) GaAs substrates with a 3° misorientation towards (111), a distinct range of $d_{\text{InP}}$ exists $\approx 0.11$ nm, where the natural tendency to develop from a high density of small QDs into several distinguishable distributions of larger QDs is observed. This transition is markedly more gradual with deposition thickness than the one observed in the molecular beam epitaxy growth of widely studied InGaAs/GaAs QDs[16, 1]. Thus the MOVPE method discussed here offers a robust well controlled method for fabrication of a suitable alternative to InGaAs structures in research of quantum effects on the nano-scale. The effects of a gradient in growth reactant concentration, CuPt-type ordering and changing the degree of misorientation of the GaAs substrate in the applied MOVPE method are also discussed.

2. Crystal growth and micro-photoluminescence studies

Growth was performed using low-pressure MOVPE in a horizontal flow quartz reactor, with two wafers exposed for each growth run. The growth temperature of the GaAs buffer and bottom GaInP layer was 700°C. Before proceeding to the deposition of InP and the GaInP capping layer, the wafer was cooled to 650°C. For the samples grown with $d_{\text{InP}} = 1.1$ nm, the temperature for the capping GaInP layer was raised to 700°C. The grown GaInP layers were nominally lattice matched to GaAs. A high InP growth rate of 0.11 nm/s was chosen.

All samples were measured at a temperature of 15K using a micro-photoluminescence ($\mu$PL) set-up. A HeNe laser with a wavelength of 543nm or 633nm was used to excite carriers in all spectra presented.

Fig.1(a) shows typical $\mu$PL spectra recorded at relatively high excitation power ($P = 15\mu\text{W}$), for QD samples grown on 3° misoriented substrates with a relatively large InP deposition thickness of $d_{\text{InP}} = 1.1$ nm. As seen for both upstream (black line) and downstream (gray line) positions, a pronounced multi-modal size distribution is observed with two broad peaks centered at 705nm and 750nm, corresponding to small and large QDs, respectively. Sharp lines corresponding to PL of individual QDs are observed in the range 725−700nm at a lower excitation power, $P = 1\mu\text{W}$, for which population of excited states in QDs is suppressed [see Fig.1(b)]. However, the peak at 750nm remains broad and featureless implying markedly higher densities of large QDs.

One method to reduce the density of large QDs is to increase the angle of misorientation on the substrate surface [12]. This reduces surface diffusion and thus inhibits QD development. Fig.1(c) shows $\mu$PL spectra recorded at low excitation power ($P = 1\mu\text{W}$) for two samples with $d_{\text{InP}} = 1.1$ nm and 0.22 nm (black and gray lines respectively) grown on (110) GaAs substrates with a 10° misorientation towards (111). As expected, it is observed that in the sample with $d_{\text{InP}} = 1.1$ nm, there is a marked suppression of the long wavelength $\mu$PL peak, but this sample also exhibits an increased density of small QDs which make this method unsuitable for this purpose.

Using the decreased misorientation angle of 3°, we show that growth may be successfully optimized to obtain a low spectral density of small QDs, combined with the tendency not to develop large QDs. As such, all further characterization discussed here was carried out on samples grown on 3° misoriented substrates.

The effect of reducing $d_{\text{InP}}$ is shown in Fig.2. The high power spectra shown in Fig.2(a)
Figure 1. Low temperature $\mu$PL spectra of InP/GaInP QDs. (a) $d_{InP} = 1.1$ nm on a $3^\circ$ off substrate, $P = 15\mu$W, showing the markedly high density of large QDs present. (b) $d_{InP} = 1.1$ nm on a $3^\circ$ off substrate, $P = 1\mu$W, showing single PL lines of small QDs. (c) Compares $d_{InP} = 1.1$ nm and $0.22$ nm on $10^\circ$ off substrates, $P = 1\mu$W, showing inhibited large QD formation and increased densities of small QDs.

are measured with $P = 5\mu$W. They show a sharp peak around 653nm, corresponding to the GaInP barrier emission. At $d_{InP} = 0.165$ nm, we measure emission from only small QDs with a narrow distribution peaked at 663nm, indicating uniform formation of very small QDs. As $d_{InP}$ is increased the QD $\mu$PL peak shifts to longer wavelength and becomes broader. Eventually at $d_{InP} = 0.33$ nm, a weak peak at 750nm is observed, corresponding to the formation of large QDs which is further enhanced for $d_{InP} = 0.44$ nm, where the emission exhibits two distinct, broad spectral QD $\mu$PL distributions.

Fig.2(b) shows $\mu$PL spectra recorded at a low excitation power ($P = 0.15\mu$W) for samples as in Fig.2(a). The optimum conditions for the growth of low density of small QDs, with a single distribution, are reached at $d_{InP} = 0.275$ nm and 0.33 nm. Over this range of $d_{InP}$, we observe a relatively small number of individual QD PL emission lines distributed in a wide spectral range, allowing clear observation of optical properties of individual QDs. Further spatial selectivity on these samples can be obtained using a standard method by fabrication of opaque metal masks with clear apertures of $\approx 1\mu$m diameter[11]. The QD density in samples with $d_{InP} = 0.275$ nm and 0.33 nm is $1\times10^9\text{cm}^{-2}$ and $8\times10^8\text{cm}^{-2}$ respectively (estimated by counting the number of lines in the spectrum at low powers). Under optimized conditions we achieve high spectral isolation of single QD PL lines with typical narrow FWHM of 0.05nm.

We note, that the observed range of $d_{InP}$ from $\approx 0.27$ nm to $\approx 0.33$ nm equivalent to 0.2 atomic monolayers (ML), which leads to growth of suitable samples at our InP deposition rate...
of 0.11 nm/s, is large in comparison to the mechanical growth-control time. This results in very reproducible growth confirmed in our further growth experiments. The variation of the dot density by a factor 1.25 in this range of $d_{InP}$ is up to a factor of 2 (30) smaller than for a similar range of deposition thicknesses in the dot density region of $\approx 10^9$ cm$^{-2}$ in molecular beam epitaxial growth of InP/GaInP (InGaAs/GaAs) QDs\cite{16, 17}.

3. Conclusions
We have realized MOVPE growth of low density InP/GaInP QDs suitable for single dot applications. The major hurdle of the presence of high densities of large QDs in the previously studied samples has been overcome. We find a reproducible, smooth transition in QD size distribution and density for varying nominal InP deposition thickness and position along the growth chamber. QD development on $3^\circ$ misorientated substrates provides a wide range of nominal InP deposition thicknesses ($\approx 0.3$ML) where conditions are suitable for single dot applications.

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