InAsP/AlGaInP/GaAs QD laser operating at ~770 nm

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Abstract. We present a study of metalorganic vapour phase epitaxy of ternary InAsP quantum dots in AlGaInP/GaAs for application in laser diodes. The properties of InAsP QD laser structures were compared with reference samples containing binary InP QDs. Based on X-ray diffraction, the molar fraction of arsenic in InAsP QDs was estimated to be ~25%. Room temperature liquid contact electro-luminescence measurements revealed a long wavelength shift of the InAsP QD emission to ~775 nm as compared with the InP QD emission at 716 nm and an increased full width at half maximum of the spontaneous emission (71 meV vs 50 meV). As cleaved, 4 mm long and 50 µm wide InAsP QD lasers operated in a pulsed regime at room temperature at ~770 nm with a threshold current density of 155 A/cm\textsuperscript{2} and a maximum output optical power of at least 200 mW. The maximum operation temperature was at least 380 K.

1. Introduction

Since the publications of Ahopelto et al [1], DenBaars et al [2] and Carlsson et al [3], self-assembled InP quantum dots (QDs) grown in (Al)GaInP matrices on GaAs substrates have attracted much interest as an alternative materials system for optoelectronic applications, primarily laser diodes [4, 5, 6], in the red – near infra-red spectral band. Recently, a significant improvement of the threshold current in InP QD laser diodes has been achieved [7, 8, 9], and other devices incorporating InP QDs such as, semiconductor disc lasers [10, 12], dual-wavelength laser diodes [12, 13], optical amplifiers with ultrafast gain dynamics [14[14]], saturable absorber mirrors for mode-locking [15, 16] and single-photon emitters [17, 18, 19, 20, 21] have been demonstrated. Also, rich nuclear spin phenomena have been observed in InP QDs [22, 23, 24, 25, 26], and efficient optical pumping of nuclear spin polarisation with ultra-long depolarisation times up to ~6000 s has been demonstrated [27]. Further to these studies, photonic crystal cavities comprising InP QDs have been fabricated [28], which would enable realisation of optically controllable spin qubits.

The operating wavelengths in the above examples can be controlled by the QD size, composition of the surrounding AlGaInP matrix and atomic diffusion between the dots and surrounding matrix, and has certain limits (here, we do not consider temperature driven spectral shift neither specific device
fabrication techniques, e.g. DFB grating [13]). In particular, a longer operation wavelength can be achieved by growing larger QDs. However, formation of misfit dislocation and difficulties with planarization of the epitaxial surface after growth of large QDs [29] limit the operation wavelength of InP QD lasers to around 750 nm at room temperature (RT).

Adding arsenic to InP QDs lowers the bandgap of the dot material and increases the lattice mismatch in respect to GaAs substrates, and, thus, offers extra flexibility in engineering the above QD structures with a potential of extending their spectral operation range towards longer wavelengths in the near infra-red band.

Growth of InAsP QDs has been attempted earlier. Vinokurov et al [30] reported InAsP QD growth by MOVPE using a mixture of arsine and phosphine but did not observe the expected long wavelength spectral shift of the QD emission, as they presumed, due to formation of smaller size InAsP QDs in comparison to the growth of nominally binary InP QDs. On contrast, Fuchi et al [31] observed a longer wavelength emission from InAsP QDs when used a droplet hetero-epitaxy technique with tertiarybutyl precursors for the group V elements and a sophisticated reagents’ switching procedure.

In this research, we applied an MOVPE process with the elements which are common for manufacturing red lasers [32, 33, 34], including commonly used group V precursors, i.e. arsine and phosphine, and trimethyl-metal precursors, for InAsP QD growth. We have demonstrated the feasibility of this approach to extend the operation of InP QD lasers towards longer wavelengths at least to ~770 nm.

2. Epitaxial growth and calibrations of P and As fractions in QDs

The epitaxy of laser structures was performed on (100) GaAs substrates with a miscut angle of 10° towards <111>A in a low pressure (150 Torr) horizontal flow reactor. The growth temperatures measured by a thermocouple inside the graphite susceptor were 710 °C or 730 °C. In addition to the groups V and III precursors mentioned in the introduction, we also used disilane and dimethylzinc for n- and p-type doping, respectively.

The active region of the laser structures consisted of 5 InP or InAsP QD sheets with a GaInP quantum well with a thickness of 8 nm grown above each QD sheet and separated by a layer of (Al0.3Ga0.7)0.52In0.48P with a thickness of 16 nm. The active region was sandwiched with an Al0.52In0.48P clad and (Al0.3Ga0.7)0.52In0.48P core waveguide with a p-GaAs contact layer on the top. Throughout the growth of the active region, the flow of phosphine was kept constant at 300 sccm, and arsine (6.25 sccm) was introduced to the reactor during growth of InAsP QDs.

To evaluate the molar fraction of arsenic in the InAsP QD layers, a calibration sample comprising of 20 periods of alternating InAsP (3.2 nm) and InP (29.2 nm) layers was grown on on-axis (100) InP substrates and analysed by a Bede QC2a X-ray diffractometer around the (004) plane. The reason for using such InP-InAsP crystalline superlattice (SL) rather than bulk InAsP layers in our calibrations was to “dilute” the average strain in the grown structures and, consequently, to avoid the complications in X-ray diffraction analysis due to the strain relaxation in the crystalline lattice.

Similarly to the InAsP QD laser active region, the calibration sample was grown at 710 °C and the same constant phosphine flow. Arsine was switched to the reactor during growth of InAsP layers. A theta-2theta diffraction scan of the calibration sample is presented in figure 1, bottom curve. The “zero” order peak of the SL is offset towards smaller diffraction angles by 216° which corresponds to a moderate compressive strain of 0.08%. Assuming no strain relaxation and abrupt reagents’ switching, the calculated diffraction spectrum (bottom curve) resulted in a good agreement with the measured one at an arsenic fraction of ~25% in the group V crystalline sublattice. The corresponding decrease of the bandgap energy of InAsP is ~250 meV.
3. Liquid contact electro-luminescence measurements

Prior to device processing, the as grown epi-wafers were examined using pulsed liquid contact electro-luminescence (LCEL). The principles of the LCEL technique for characterization of laser diodes were formulated and realized by Zory et al [35, 36, 37]. In our earlier work [38], we have successfully used LCEL for characterization of InP QD lasers. The setup used in the current research was built around a polaron cell where the light emitted through the top p-type GaAs cap layer was collected into an optical fibre led to an Ocean Optics spectrometer. The pulse duration (1 ms) and current (0.5-80 mA) were kept minimal to avoid formation of bubbles of hydrogen in the cell which prevent current transport. Also, the LCEL measurements under the above excitation parameters caused just minimal surface damage to the cap layer and did not affect the subsequent device fabrication process.

The LCEL spectra of InP and InAsP QD samples are presented in figure 2. The emission spectrum of nominally binary InP QDs was dominated by an emission centred at 716 nm with a full width at half maximum (FWHM) of 20.6 nm (equivalent to ~50 meV). Adding arsine to the reactor during the QD growth resulted into a wavelength shift of the QD emission to 775 nm, a decreased peak intensity and an increased FWHM of 34.3 nm (~71 meV). The latter indicates a higher degree of compositional and size inhomogeneity of InAsP QDs. It is important to mention that the long wavelength shift of ~60 nm (132 meV) of the InAsP QDs is considerably smaller than expected from the arsenic fraction derived from the X-ray diffraction data and the corresponding bandgap reduction of InAsP. This discrepancy can be explained by a smaller average InAsP QD size in comparison to InP QDs, as suggested by Vinokurov et al [30].

4. Transmission electron microscopy (TEM)

We performed TEM studies on the QD laser samples to confirm the QD formation and assess the QD size and density. Representative TEM images are shown in figure 3. In both samples, the QDs are aligned in isolated vertical stacks with a density of the order of $10^9 - 10^{10}$ cm$^{-2}$. The InP QDs generally
appear larger and the stacks more regular than the InAsP QDs, with a height of 4-5 nm and a lowermost dot diameter of 40-50 nm, whereas the InAsP dots have a height of 2-3 nm, a lowermost dot diameter of 20-30 nm and greater variability in dot diameter through the stack. It is not yet clear whether the smaller dot size and greater size variation is a fundamental property of the InAsP dots (e.g. due to a higher lattice mismatch) or simply results from using growth conditions optimised for InP dots, which may not be optimal for the InAsP dots. However, the observed greater variability and smaller average size of the InAsP QDs are in a good agreement with their luminescence properties described in the previous section.

Figure 2. LCEL spectra of as grown InP and InAsP QD laser wafers at RT.

Figure 3. Transmission electron microscope images (bright field, g = 004) of InAsP QD (upper image) and InP QD (lower image) laser structures.

5. Laser results
The epitaxial wafers were processed in 4 mm long, 50 μm wide, oxide isolated stripe lasers with uncoated facets. The lasers were operated in pulsed mode (1 kHz, 1000 ns) to reduce self-heating. Laser oscillations of InP and InAsP QD samples were observed at around 720 nm and 770 nm (figure 4), respectively, close to the corresponding LCEL wavelengths. Both samples delivered optical powers of at least 200 mW.

The results of detailed measurements of the temperature dependence of the threshold current densities are presented in figure 5. They confirmed higher threshold current densities for InAsP QD samples over the studied temperature interval. However, the both structures showed reasonably small threshold current densities (e.g. 123 A/cm² and 155 A/cm² at RT for InP and InAsP QD samples, respectively) and lased up to at least 380 K. At elevated temperatures, above ~350 K, the threshold current density of the InAsP QD laser exhibits a trend of a slightly slower increase as compared to that.
of the InP QD laser due to the greater confinement potential in InAsP QDs. In general, the performances of InP and InAsP QD lasers compare well with those of uncoated and unmounted lasers based on other QD and quantum well materials systems operating around the same spectral range of ~700-800 nm [39, 40, 41, 42].

Detailed characterization and analysis of the laser properties of InAsP QDs can be found elsewhere [43].

![Emission spectra of InP and InAsP QD laser diodes just above the threshold.](image1)

![Temperature dependence of the threshold current densities of InP and InAsP QD laser diodes.](image2)

**Figure 4.** Emission spectra of InP and InAsP QD laser diodes just above the threshold.  
**Figure 5.** Temperature dependence of the threshold current densities of InP and InAsP QD laser diodes.

6. **Conclusions**

We have demonstrated MOVPE of InAsP QDs as a feasible approach to extend the operation range of InP QD lasers towards longer wavelengths. A long wavelength shift of the QD spontaneous emission of ~60 nm was achieved at a molar As fraction of ~25% in the group V sublattice. In comparison to InP QDs, InAsP QDs shows a greater size and composition variability as evident from TEM images and increased inhomogeneous broadening of the LCEL spectra. Uncoated, 4mm long and 50 µm wide stripe InAsP QD laser operated at ~770 nm in a pulse regime with a threshold current of 155 A/cm² and maximum optical powers of at least 200 mW at RT and maximum operation temperature of at least 380 K.

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