Formation of defect-free InGaAs-GaAs quantum dots for 1.3 $\mu$m spectral range grown by metal-organic chemical vapor deposition

I. L. Krestnikov†, N. N. Ledentsov†, M. V. Maximov‡, D. Bimberg†, D. A. Bedarev‡, I. V. Kochnev‡, V. M. Lantratov‡, N. A. Cherkashin‡, Yu. G. Musikhin‡ and Zh. I. Alferov‡
† Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany
‡ Joffe Physico-Technical Institute, St Petersburg, Russia

Recently, remarkable progress is reported in GaAs-based quantum dot (QD) lasers emitting near 1.3 $\mu$m [1–5]. For single-sheet QD lasers ultralow threshold current densities ($\sim$20 A/cm$^2$, 300 K) are reported for the case of low mirror losses [3]. To come to practical device applications requesting high power and high-frequency applications one needs, however, to use stacked 1.3 $\mu$m-emitting QDs [2] to ensure sufficient modal gain to maintain lasing at requested cavity lengths and facet reflectivities. Due to this approach, high-power (2.7 W CW) operation [5] in broad area devices and single transverse mode (300 mW) operation in narrow (7 $\mu$m) stripes [4] were realized. Very recently, modal gain as high as 50 cm$^{-1}$ in the 1.3 $\mu$m range was obtained for injection lasers based on 10-fold stacked InAs QDs in a GaAs matrix [6].

All these results are obtained, however, for long-wavelength QDs grown by molecular beam epitaxy (MBE) [7]. As opposite, despite the first GaAs-based QDs emitting in the 1.3 $\mu$m range both under photoexcitation [8] and current injection [9] were reported for MOCVD growth, these structures seemed to be not suitable for laser applications due to low density of QDs and high density of defects [9–10] and only lasers emitting in the range of up to 1.1 $\mu$m are developed [11–12].

In this paper we have grown by metal-organic chemical vapor deposition structures with InGaAs QDs and studied the influence of post-deposition treatment on their structural and optical properties. The samples studied in this work are grown by MOCVD using trimethyl gallium (TMG), trimethyl aluminium (TMA), ethyl-dimethyl indium (EDMI) and arsine (AsH$_3$). Hydrogen is used as a carrier gas. The total pressure in the reactor is kept at 76 Torr and the flux ratio between group III elements and arsine is 75. The structures are grown on semi-insulating GaAs (100) substrates. First, a $\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}$ buffer layer (0.8 $\mu$m) is grown at 600°C, followed by a 0.1 $\mu$m-thick GaAs layer. The substrate temperature is then reduced to 490°C and $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ deposition (or $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ multilayer deposition) is performed. In the first case 3 monolayers (ML) of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ are deposited. In the second case the deposition of 3 MLs of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ is followed by multi-cycle $\text{GaAs}$ (140 nm) — $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ (2.5 ML) deposition (two periods). After this 40 nm of GaAs is grown at 490°C, the substrate temperature is increased to 600°C, and 40 nm of GaAs is deposited. To avoid surface recombination of nonequilibrium carriers 20 nm of $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ is grown on the top at the same temperature. As opposite to the case of vertically-coupled MOCVD QDs [13], large separation between QD prevented any wavefunction-coupling effects. Two types of structures were grown. In one case, the InGaAs deposition was completed with GaAs overgrowth at 490°C. In the other case, for both single and multi-sheet structures, first thin GaAs layer (8 nm) is deposited at 490°C and,
then the substrate temperature is increased to 600°C for 10 minutes under AsH₃ exposure. During this procedure InAs accumulated in large clusters, which are not covered by thin GaAs cap layer, evaporates and redistributes through the GaAs surface forming the second wetting layer. This procedure was shown using deep level transient spectroscopy (DLTS) to result in complete suppression of the EL2 and EL3 traps, associated with dislocations, and in reduction of the concentration of deep traps associated with point defects by more than one order of magnitude [14-1.

Transmission electron microscopy (TEM) and high-resolution electron microscopy (HREM) studies are performed by using a high voltage Philips EM-420 (100kV) microscope. Plan-view and cross-section TEM images are taken under (220) and (200) diffraction conditions, respectively. Photoluminescence (PL) is excited by using the 514.5 nm line of a Ar⁺ laser and detected by using a cooled germanium pin-photodetector.

In Fig. 1 we show plan view TEM images of the structures with single QD insertions grown without or with annealing steps (Fig. 1(a) and (b), respectively). One can clearly see coherent InGaAs QDs on both images with areal density of about 2 × 10¹⁰ cm⁻². The lateral size of QDs is about 20–25 nm, being similar to reported in [13]. In addition to coherent QDs, the structure grown without the annealing step demonstrates contrast features due to dislocated InGaAs clusters, having a density of about 0.5–1 × 10⁹ cm⁻². No such clusters are revealed in the structure grown with the annealing step.

Plan view (a, c) and cross-section (b, d) TEM images of the structures with 3-fold stacked InGaAs insertions grown without (a, b) and with (c, d) annealing steps are shown in Fig. 2. In the case of the plan-view images, the foil thickness was kept below 100 nm to allow QD imaging. The only upper row of InGaAs QDs is trapped in the TEM foil in this case. As it is clearly seen from Fig. 2, the QDs are present in the upper sheet with InGaAs insertion only in the case of the structure grown with the annealing steps. The QD in-plane density in this case is similar to that for the single-sheet insertion (~2 × 10¹⁰ cm⁻²). As opposite, the structure grown without the annealing steps demonstrates complete disappearance of QDs in the upper InGaAs sheet region. One may conclude, that most of the InGaAs material is trapped in the regions in the vicinity of propagating dislocations originating at the clusters in the first row with QDs. These dislocations are clearly visible in the plan-view (Fig. 2(a)) cross-section TEM (Fig. 2(b)) images of the stacked InGaAs insertions grown without the annealing step. Thus, the concept of stacking of QDs does not work without the annealing step.

Cross-section TEM image of the structure grown with the annealing step demonstrates
complete suppression of defect formation (see Fig. 2 (c),(d)). Higher magnification cross-section TEM images of this sample demonstrate also an additional dark stripe in the contrast due to the second InGaAs wetting layer formed from the InGaAs accumulated in large dislocated clusters, which are not completely covered with 8 nm-thick GaAs cap layer, as opposite to coherent QDs having smaller height [8, 10, 13].

Formation of defects has significant impact on optical properties of the structures studied. The PL spectrum of the sample with single InGaAs QD insertion grown with the annealing step is shown in Fig. 3. At room temperature relatively broad PL emission in the range 1–1.4 μm peaking at around 1.3 μm is observed. At high excitation density PL
due to excited state of quantum dots becomes more pronounced. The peak $I$ is attributed to InGaAs two-dimensional islands having several monolayer height similar to discussed in [10, 11]. The single-sheet structure grown without the annealing step shows similar PL spectrum, but the PL intensity is an order of magnitude lower at 300 K.

In the case of the stacked QD structure grown with the annealing step the PL spectrum is similar to shown in Fig. 3 in lineshape and intensity both at low temperatures and at 300 K. As opposite, the PL spectrum of the stacked InGaAs-GaAs structure grown without the annealing step shows only weak emission in the 850–1000 nm range at low temperatures and essentially no PL in the long-wavelength range. This indicates that most of the carriers are trapped in the wetting layer- or island-induced states dominating in the upper rows of the stacked structure in agreement with TEM data. At room temperature, the PL intensity degrades by 3 orders of magnitude in this case.

To conclude, we have investigated structural and optical properties of MOCVD-grown single and stacked InGaAs insertions formed with and without annealing step after thin-layer overgrowth. We found that this overgrowth/annealing procedure results in elimination of dislocated InGaAs clusters, thus, only the structures grown with such an approach are suitable for long-wavelength laser applications for the growth mode chosen in this work. Additionally, according to our results, growth of stacked long-wavelength QDs is principally possible only when QDs are subjected to the cluster-elimination procedure.

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References