Room-temperature continuous-wave lasing from stacked InAs/GaAs quantum dots grown by metalorganic chemical vapor deposition

F. Heinrichsdorff,[a] M.-H. Mao, N. Kirstaedter, A. Krost, and D. Bimberg
Institut für Festkörperphysik, Technische Universität Berlin, 10623 Berlin, Germany

A. O. Kosogov[b] and P. Werner
Max-Planck-Institut für Mikrostrukturphysik, D-06120 Halle, Germany

(Received 9 December 1996; accepted for publication 1 May 1997)

We report on quantum dot (QD) lasers made of stacked InAs dots grown by metalorganic chemical vapor deposition. Successful growth of defect-free binary InAs/GaAs QDs with high lateral density ($d_l \geqslant 4\times10^{10}$ cm^{-2}) was achieved in a narrow growth parameter window. The room-temperature photoluminescence (PL) intensity is enhanced up to a factor of 3 and the PL peak width is reduced by more than 30% when a thin layer of $In_{0.3}Ga_{0.7}As$ is deposited onto the InAs QDs. A QD laser with a single sheet of such InAs/InGaAs/GaAs QDs exhibits threshold current densities as low as 12.7 and 181 A/cm^2 at 100 and 300 K, respectively. Lasers with threefold stacked QDs show ground-state lasing and allow for cw operation at room temperature. © *1997 American Institute of Physics.* [S0003-6951(97)01727-0]

Since the theoretical prediction of superior properties of semiconductor lasers with quantum dots (QDs) serving as the active medium,[1,2] as compared to quantum wells (QWs), it has taken a decade until the first quantum dot semiconductor laser was realized[3] by means of molecular beam epitaxy (MBE). Meanwhile, several reports on lasers with the active region made of single and multiple In(Ga)As QD sheets in a GaAs matrix have been published by various groups.[4–7] The use of vertically stacked QD layers is beneficial for laser applications, since the increased effective number of dots increases the modal gain and helps to overcome gain saturation.[8] Furthermore, for electronically coupled dots, the localization energy and capture efficiency may be enhanced as compared to single dot layers.[9] A QD laser with a room-temperature (RT) threshold current density as low as 62 A/cm^2 for four-side-cleaved samples has been produced[10] with a threefold stack of vertically coupled QDs, even though the size distribution induced energetic broadening of the QD emission is still far from being optimum.

All the above-mentioned reports on QD lasers deal with structures grown by MBE. In contrast, there exist only two very recent publications concerning QD lasers grown by metalorganic chemical vapor deposition (MOCVD). A laser with $In_{0.5}Ga_{0.5}As$ dots as active material has been presented by Maximov *et al.*[11] By inserting a single dot sheet in a GaAs/AlGaAs QW, a very high T_o of 385 K up to 330 K was reached. Schur *et al.* recently reported on ternary InGaAs/GaAs QDs in a vertical microcavity structure that showed stimulated emission under optical excitation at 77 K.[12] In both papers, single sheets of ternary InGaAs served as the dot material, since both the use of pure InAs and the attempt to stack several QD layers were observed to lead to the formation of large dislocated clusters in MOCVD.[13]

In this letter, we report on a successful fabrication of high-density defect-free binary InAs dots by MOCVD. The optical quality of the QDs is enhanced by overgrowth of the InAs QDs with a thin ($d=1.4$ nm) $In_{0.3}Ga_{0.7}As$ layer. The absence of defects allows for the growth of device quality stacked dot layers. Transmission electron microscopy (TEM) images reveal pronounced vertical ordering of the QDs. Low threshold separate confinement heterojunction lasers with single and stacked QD sheets serving as the active medium operating at room temperature are demonstrated.

The samples were grown at a 20 mbar total reactor pressure with TMIn, TMGa, TMAl, DMZn, SiH_4 (5% in H_2), and pure AsH_3 as source materials on exactly oriented (001) $\pm0.1°$ GaAs:Te substrates. After deposition of the InAs at a growth temperature (t_{Gr}) of 485–505 °C and a growth rate of 1.5–4.5 ML/s, the growth was interrupted for 8 s to form the QDs. During this growth interruption (GRI), the AsH_3 flux was switched off. For the InAs deposition, the V/III ratio was as low as 25–40. In addition to the InAs QDs, we produced combined binary/ternary InAs/InGaAs QDs (BTQDs) by deposition of a thin layer of $In_{0.3}Ga_{0.7}As$ ($d=1.4$ nm) onto the InAs QDs before growth of the GaAs cap layer. For these novel QDs, two GRIs of 4 and 8 s duration without AsH_3 were applied after InAs and InGaAs deposition, respectively.

For all laser devices, similar structures consisting of either single or stacked QD sheets introduced in the center of a 150 nm thick GaAs region embedded between two $Al_xGa_{1-x}As$ ($x=0.5$–0.7, $d=0.8$–$1\ \mu$m) layers were used. On top of the lower (and below the upper) AlGaAs cladding layers, an AlGaAs/GaAs superlattice (20×2 nm/2 nm) was introduced to provide a smooth surface for the QD growth. Without the superlattices, we found a reduction of the photoluminescence (PL) efficiency for QDs grown on thick AlGaAs layers as compared to QDs grown without underlying AlGaAs that is attributed to surface roughening during AlGaAs deposition.[14] On top of this structure a p^+-GaAs contact layer of 600 nm thickness was grown. The laser diodes were fabricated in a shallow stripe geometry of 8–50 μm in width and 0.5–2.0 mm in length.

For photoluminescence measurements, the samples were excited with the 514 nm line of an Ar^+ laser and the luminescence light was detected with a LN_2 cooled germanium PIN diode. For electroluminescence (EL) measurements, ei-

[a]Electronic mail: franki@w422zrz.physik.TU-Berlin.DE
[b]Also at Institut für Festkörperphysik, Technische Universität Berlin.

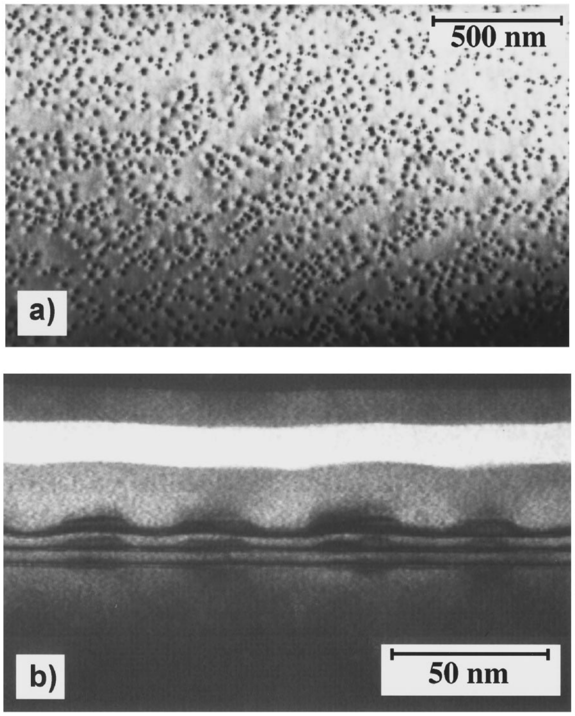

FIG. 1. Plan-view (a) and cross-section (b) TEM images of triple InAs/GaAs dot stacks with 4 nm separation layer thickness.

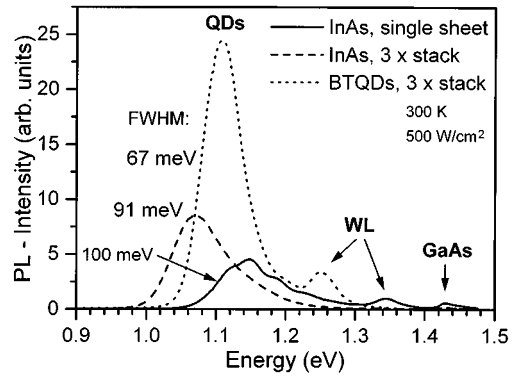

FIG. 2. Room-temperature PL spectra of single InAs QD sheet and threefold stacks of InAs QDs and BTQDs.

ther cw or pulsed mode current injection with a repetition frequency of 5 kHz and pulse width of 500 ns was used. Plan-view and cross-section TEM images of conventionally prepared samples were taken with JEOL JEM1000 (1 MV) and JEOL JEM4000 (400 kV) microscopes.

Figure 1(a) shows a large scale plan-view TEM image ($g=220$) of a threefold stacked InAs QD sample. The integral dot density is 4×10^{10} cm^{-2}. Locally, however, the density reaches much higher values in domainlike dot agglomerations. We did not find any macroscopic defect for a sample area of more than 25 μm^2. A higher magnification (002) dark field cross-section image of the sample is shown in Fig. 1(b). Besides the threefold QD stack (dark contrast) and the GaAs matrix, the $Al_{0.25}Ga_{0.75}As$ cap layer (25 nm above the dots) is visible in white contrast. A perfect vertical alignment of the dots can be seen. Each QD column produces a pronounced bending of the lower AlGaAs/GaAs interface. This corrugation is partly smoothed out in the AlGaAs layer. Thus, the sample surface appears nearly flat. The lateral size of the stacked dots, as deduced from the cross-section image, increases from 9 to 12 nm in the first QD sheet to 15–18 nm in the third one. The exact geometry of the dots cannot be clearly seen from the image due to the impact of strain fields on the image contrast. From higher magnification plan-view images, we found the stacked dots' bases to be of rhombic shape (near to square), similar to the findings for ternary InGaAs QDs.[15,16]

In Figure 2, RT PL spectra of a single QD layer and threefold stacks of InAs QDs (d_{sep}=4 nm) and BTQDs (d_{sep}=21 nm) under excitation of 500 W/cm^2 are shown. All spectra are dominated by the broad QD luminescence at 1.07–1.15 eV. An additional weaker line around 1.35 eV for the InAs QDs and 1.25 eV for the BTQDs can be seen, which is attributed to the luminescence from the wetting layer. At 1.43 eV, a GaAs related luminescence peak is also present. It should be pointed out that for InAs layers with a thickness reduced by 20%, as compared to the samples shown in Fig. 2, we do not find any QD luminescence but only a quantum well PL peak around 1.35 eV. If the nominal InAs thickness is further increased by 10%, the QD luminescence intensity drops by more ~30% due to the onset of plastic strain relaxation.

A comparison of single and stacked dot InAs QD spectra shows a redshift of the dot related luminescence by ~70 meV and a significant enhancement of the PL efficiency for the latter. The wetting layer (WL) peak nearly vanishes for the stacked sample and is only slightly redshifted by 24 meV, thus, enhancing the energetic separation of carriers in the dots and in the wetting layer. In contrast, for the BTQD stack, the wetting layer peak is redshifted by 100 meV, but the QD peak remains nearly unshifted. The BTQD PL peak width is reduced by one third as compared to that of the threefold InAs QD stack. Surprisingly, the RT PL intensity of the BTQDs is enhanced as compared to the binary InAs QDs, although a reduced energetic separation between QDs and the WL is normally associated with an enhanced carrier evaporation from the QDs at elevated temperatures. Possible origins for the increase in PL intensity are an improved overall quantum efficiency for the BTQD samples, as indicated by the enhanced WL peak intensity, and a more efficient carrier capture into the BTQDs, as compared to the InAs QDs. The PL peak position of the threefold BTQD stack is identical to that of a single BTQD sheet (not shown here), since the thick separation layer (d_{sep}=21 nm) prevents any electronical or structural coupling of subsequent layers. In contrast, the thin separation layer of the InAs QD stack (d_{sep}=4 nm) leads to an influence of the lower lying QD sheets on the size/shape of the upper QDs. Thus, the redshift of the PL peak from the stacked QDs is attributed to the increased average QD size, as shown in Fig. 1(b) and probable electronic coupling of subsequent QD sheets.

In view of lasing, the BTQDs also demonstrate much superior performance. From lasers with fivefold InAs QD stacks (d_{sep}=4 nm), we derived RT threshold current densities j_{thr}=1.3 kA/cm^2 under pulsed mode current injection. In contrast, the j_{thr} values measured for a single sheet BTQD laser are as low as 12.7 A/cm^2 at 100 K and 181 A/cm^2 at 293 K (0.05×2 mm^2 stripes). For a threefold BTQD stack,

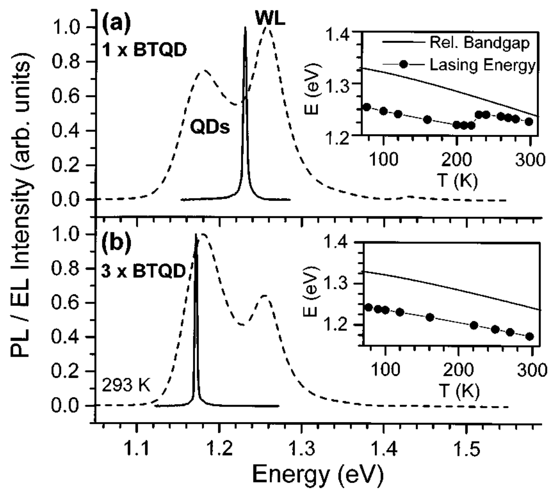

FIG. 3. Room-temperature EL spectra (straight) under pulsed mode current injection at $I=1.05I_{\mathrm{thr}}$ and PL spectra (dashed) of single sheet (a) and triple stack BTQD laser (b). Temperature dependence of the lasing energy as compared to the relative InAs band gap is given in the insets.

we obtained j_{thr} values of 21.7 A/cm^2 (100 K) and 220 A/cm^2 (293 K). The RT EL and PL spectra ($I_{\mathrm{exc}}=500$ W/cm^2, top cladding etched off) for the BTQD lasers are shown in Fig. 3.

A comparison of the QD PL peak energies in Fig. 3 to that in the BTQD spectrum of the test structures in Fig. 2 yields a blueshift of 73 meV for the laser structures. This shift can be explained by thermally induced intermixing of the QDs with the surrounding GaAs during growth of the upper cladding and contact layers ($T_{\mathrm{Gr}}=640\,°\mathrm{C}$).[17,18] The stacked QD laser emits at the low-energy tail of the PL peak, indicating ground-state lasing for up to RT. In contrast, the lasing emission of the single sheet QD laser is observed on the high-energy tail of the QD PL peak. Kirstaedter *et al.* assigned this effect to an enhanced contribution of excited states to the lasing activity due to gain saturation of the ground state.[19] This explanation is confirmed by the temperature dependence of the laser energies as compared to the relative InAs band-gap shift[20] shown in the insets of Fig. 3. The lasing energy of the single sheet laser shows a sudden increase of 31 meV at 230 K, similar to the observations of Shoji *et al.*[21] For the stacked QD laser, no such blueshift is observed, since the higher number of QDs increases the ground-state modal gain.[8] The higher modal gain of the threefold QD stack allows for room-temperature cw operation. For a stripe geometry of 8 μm$\times$500 μm, we measured a threshold current of 24 mA and an emission energy of 1.188 eV.

In summary, we have grown device quality single sheet and stacked binary InAs/GaAs quantum dots by low-pressure MOCVD. By combining InAs and $In_{0.3}Ga_{0.7}As$ deposition for the QDs, we achieved an increase of the QD's luminescence efficiency by a factor of 3. QD lasers with single sheet and stacked QDs have been produced. Single sheet QD lasers exhibit threshold current densities of 12.7 and 181 A/cm^2 at 100 and 300 K, respectively. Lasers with threefold stacked QDs show laser emission on the QDs ground state and allow for RT cw operation.

The authors acknowledge K. Schatke for expert technical assistance during the MOCVD growth and M. Kuttler and M. Strassburg for performing secondary-ion-mass spectroscopy measurements. This work is funded by the DFG in the framework of SFB 296.

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