InAs quantum dots grown by MOCVD in GaAs and metamorphic InGaAs matrices

R A Salii¹, N A Kalyuzhnyy¹,², N V Kryzhanovskaya², M V Maximov², S A Mintairov¹,²,³, A M Nadtochiy²,³, V N Nevedomskiy¹ and A E Zhukov²

¹ Ioffe Institute, Saint Petersburg, Russia
² St Petersburg Academic University, St Petersburg, Russia
³ Solar Dots Ltd, St Petersburg, Russia

e-mail: r.saliy@mail.ioffe.ru

Abstract. We have studied MOCVD-grown structures with InAs layers in GaAs and metamorphic InGaAs matrices. Deposition of 2 ML of InAs in a GaAs or In₀.₂₅Ga₀.₇₅As matrix results in quantum dots formation with photoluminescence (PL) peaks at 1240 and 1380 nm, respectively. In case of deposition of 2 ML of InAs in a In₀.₃₀Ga₀.₇₀As metamorphic matrix, formation of a corrugated quantum well emitting at 1450 nm has been revealed. The integrated PL intensity of the metamorphic structures is much higher than that for the GaAs based structure. For the metamorphic structures, the thermal escape of carriers from InAs quantum dots (a quantum well) results in high intensity of the PL line due to InGaAs matrix at room temperature.

1. Introduction
Quantum size objects such as quantum wells (QWs), quantum wires and quantum dots (QDs) provide significant improvement of the performance of semiconductor devices [1]. Using InAs QDs with pseudomorphic QW capping in a GaAs matrix has allowed obtaining the lowest laser threshold currents [2-4]. The QWs and QDs have also found an application in photovoltaic converters (PVC) providing improvements of their parameters [5-8]. Among other advantages, QWs and QDs enable achieving a longer emission wavelength as compared to bulk materials. This becomes especially apparent in case of QDs. For instance, the use of InAs/InGaAs QDs in a GaAs matrix has provided low threshold lasing in the range of 1250 - 1350 nm.

One of the promising ways of further wavelength increase, e.g. towards a 1.55 μm telecom window, is the use of a metamorphic InGaAs matrix for epitaxial structures on GaAs substrates [9]. MBE-grown lasers emitting in the 1.3 - 1.55 μm interval have been created using InAs QDs in the InGaAs matrix grown on a GaAs substrate via a metamorphic buffer [10, 11]. Metamorphic buffers have also been successfully used for PVC [12-14]. However, most of the research in the field of QDs has been carried out using molecular beam epitaxy (MBE), whereas at the moment the metal-organic chemical vapor deposition (MOCVD) has become the main production technology for epitaxy of various semiconductor devices.

In the present work, we study InAs QD formation by MOCVD in a lattice matched GaAs matrix as well as in a metamorphic InGaAs matrix.
2. Experimental details

Structures under study were grown using a horizontal MOCVD reactor under 100 mbar on GaAs (100) substrates. Trimethylgallium, trimethylaluminum and trimethylindium were used as III-group atom precursors. Arsine was used as a source of As atoms.

We have grown and studied three structures (figure 1). In all the structures, the active region was formed by the deposition of 2 monolayers (ML) of InAs. The InAs layer was centered in the 500 nm-thick InₓGa₁₋ₓAs matrix. (AlₓGa₀.₅)₁₋ₓInₓAs wide-bandgap layers were used as barriers preventing carrier diffusion from the matrix to the substrate and structure surface. Structure N1 was made lattice matched to the GaAs substrate ($x = 0$) (figure 1(a)), whereas for structure N2 $x = 24\%$ (figure 1(b)) and for structure N3 $x = 30\%$ (figure 1(c)). The details of the metamorphic buffer design are presented elsewhere [13].

Transmission electron microscopy (TEM) study was carried out with a JEOL JEM 2100F microscope at 200 kV accelerating voltage. The samples were prepared by preliminary mechanical thinning followed by final ion beam milling (3.5 keV Ar⁺ ions).

Photoluminescence (PL) spectra were measured using a 532nm Nd:YAG laser with an output power of 350mW. PL was focused on a monochromator. All measurements were carried out by means of a cooled Ge photodetector using a standard lock-in amplifier.

![Figure 1](image_url). Schematic of the structure N1 (a), structure N2 (b) and structure N3 (c).

3. Results and discussion

TEM studies revealed formation of self-assembled Stransky–Krastanov (SK) QDs for structure N1 and structure N2 (figure 2(a), (b)), whereas for structure N3 only a corrugated thin InAs layer was observed (figure 2(c)). The latter result can be explained by the insufficient InAs thickness in case of using the In₀.₃₀Ga₀.₇₀As matrix. It has been shown [8], that for InAs QDs grown by MOCVD in a GaAs matrix, the SK QDs with sufficiently high crystal quality are formed in the 1.4 – 2.4 ML range. The critical thickness necessary for coherent QDs formation due to elastic strain relaxation is 1.4 ML. The thickness of QDs coalescence with formation of large defected InAs islands is 2.4 ML. Increasing the In content in the metamorphic matrix leads to a reduction of the mismatch of lattice constants of the matrix and InAs. As a result, the critical thickness increases. Thus, one can conclude that the critical thickness exceeds 2ML in case of the In₀.₃₀Ga₀.₇₀As matrix. The surface density of the SK QDs grown
in the $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ is $\sim 10^{11}$ cm$^{-2}$ (figure 2(b)) and seems to be higher than that for the GaAs matrix (figure 2(a)).

For structure N1 the PL spectrum under low excitation power at 300 K shows two peaks at 1140 and 1240 nm which correspond to QDs of different sizes or ground-state and excited-state optical transitions (figure 3(a)). The bi-modality of the MOCVD grown InAs QDs in GaAs matrix has been discussed elsewhere [8, 15]. For structure N2 a broad peak centered at 1380 nm is observed (figure 3(a)). We conclude that this peak corresponds to a recombination via SK QDs shown in figure 1(b). Besides this peak, a narrower peak at 1140 nm equal to the bandgap of the $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ matrix is observed. Although the QDs were not observed in the TEM pictures of structure N3 (figure 1(c)), its PL spectrum looks similar to that for structure N2 (figure 3(a)). The long wavelength PL peak corresponding to recombination via the InAs layer is observed at 1450 nm. The large spectral width of this line can be explained by the strong corrugation and non-uniformity of the InAs layer. The spectral position of the second narrower peak (1240 nm) is equal to the $E_g$ of $\text{In}_{0.30}\text{Ga}_{0.70}\text{As}$.

![Figure 2](image)

**Figure 2.** Bright-field cross-section TEM micrographs of QD regions for structure N1 (a), structure N2 (b) and structure N3 (c).

The presence of the matrix peak at the PL spectra for structure N2 and structure N3 can be explained by enhanced thermal escape of the carriers from InAs into the matrix. Indeed, the difference between the energies of the QDs and the matrix bandgaps for structure N2 ($\sim$ 180 meV) is two times lower than that for structure N1 ($\sim$ 440 meV). For structure N3, this difference is $\sim$ 140 meV. This
explanation is also confirmed by the PL data taken at 77 K (figure 3(b)), which do not reveal matrix peaks for all the structures.

At 77 K, the integrated intensities of the PL spectra for metamorphic structures N2 and N3 are several times higher in comparison with GaAs-based structure N1, whereas at 300 K the intensities of all three structures are comparable. This can be explained by non-radiative recombination on defects in metamorphic structures. As the defects capture cross section decreases with decreasing temperature, their impact on the PL spectra at 77 K becomes negligible.

![Figure 3](image)

**Figure 3.** PL spectra for structure N1 (red), structure N2 (green) and structure N3 (blue) under low excitation power at: (a) – room temperature, (b) – 77 K.

Under high excitation density, the PL spectrum of structure N1 shows three additional peaks at 1040, 930 and 860 nm corresponding to the QDs excited state, QDs wetting layer and GaAs matrix, respectively (figure 4(a)). At the same time, the shape of the PL spectra of structures N2 and N3 do not significantly change but their integrated intensities become much higher in comparison with that of structure N1 (figure 4(a)). We suggest that this finding can be explained by saturation of the non-radiative recombination channels for metamorphic structures under high excitation density.

At 77 K under high excitation density, all the spectra show a matrix peak (figure 4(b)) attributed to saturation of the recombination via QDs. Under such conditions, the metamorphic structures also show a higher integrated intensity of the PL as compared to structure N1.

![Figure 4](image)

**Figure 4.** PL spectra for structure N1 (red), structure N2 (green) and structure N3 (blue) under high excitation power at: (a) – room temperature, (b) – 77 K.

### 4. Conclusion

The 2 ML-thick InAs insertions were epitaxially deposited by MOCVD into GaAs, metamorphic In\textsubscript{0.24}Ga\textsubscript{0.76}As or metamorphic In\textsubscript{0.30}Ga\textsubscript{0.70}As matrices. TEM studies have shown that in case of GaAs
and In$_{0.24}$Ga$_{0.76}$As matrices, the formation of SK QDs takes place, whereas for In$_{0.30}$Ga$_{0.70}$As, the InAs thickness is insufficient for QDs formation. The PL and TEM data has allowed to conclude that the density of QDs in In$_{0.24}$Ga$_{0.76}$As is higher than that for GaAs. It has also been found that a smaller difference between the bandgap energy of InAs QDs or a QW and a matrix for metamorphic structures results in carrier thermal escape from the active region into the matrix with subsequent recombination there.

Acknowledgments
The study was supported by the Russian Foundation for Basic Research (grant #16-29-03127). Structural characterization by TEM was made on the equipment of the Federal Joint Research Centre «Material science and characterization in advanced technology» (Ioffe Institute, St. Petersburg, Russia).

References
[1] Bhattacharya P, Fornari R and Kamimura H 2011 Comprehensive Semiconductor Science and Technology, Six-Volume Set, 1st Edition ELSEVIER S & T
[2] Kirstaedter N, Ledentsov N N, Grundmann M, Bimberg D, Ustinov V M, Ruvimov S S, Maximov M V, Kop’ev P S, Alferov Zh I, Richter U et al. 1994 Electron. Lett. 30 1416
[3] Lester L F, Stinz A, Li H, Newell T C, Pease E A, Fuchs B A and Malloy K J 1999 IEEE Photon. Technol. Lett. 11 931
[4] Maximov M V, Zhukov A E Ed. S E Lyshesvski 2014 Quantum Dot Lasers, In Dekker Encyclopedia of Nanoscience and Nanotechnology, Third Edition. CRC Press: New York, 3986–4000.
[5] Browne B, Lacey J, Tibbits T, Bacchin G, Wu T-C, Liu J Q, Chen X, Rees V, Tsai J and Werthen J-G 2013 AIP Conference Proceedings1556 3-5
[6] Fujii H, Toprasermpong T, Wang Y, Watanabe K, Sugiyama M and Nakano Y 2014 Prog. Photovolt: Res. Appl. 22 784–795
[7] Wheeldon J F, Valdivia C E, Masson D, Proulx F, Riel B, Puetz N, Desfonds E, Fafard S, Rioux B, Spring Thorpe A J, Arêz R, Aimez V, Armstrong M, Swinton M, Cook J, Shepherd F, Hall T J and Hinzer K 2010 Proc. SPIE 7750 77502Q
[8] Kalyuzhnyy N A, Mintairov S A, Salii R A, Nadtochiy A M, Payusov A S, Brunkov P N, Nevedomsky V N, Shvarts M Z, Marti A, Andreev V M and Luque A 2016 Prog. Photovolt: Res. Appl. 24 1261–1271
[9] Wang Sh 2012 Lattice Engineering - Technology and Applications CRC Press
[10] Semenova E S, Zhukov A E, Mikhrin S S, Egorov A Yu, Odnoblyudov V A, Vasil’ev A P, Nikitina E V, Kovsh A R, Kryzhanovskaya N V, Gladyshev A G et al. 2004 Nanotechnology 15 S283 - S287
[11] Karachinsky L Ya, Kettler T, Novikov I I, Sharunov I K, Kryzhanovskaya N V, Zhukov A E, Semenova E S, Vasil’ev A P et al. 2006 Semiconductor Science and Technology 21, 691-696
[12] Sasaki K, Agui T, Nakaïdo K, Takahashi N, Onitsuka R and Takamoto T 2013 AIP Conf. Proc. 1556 22
[13] Mintairov S A, Emelyanov V M, Rybalchenko D V, Salii R A, Timoshina N K, Shvarts M Z and Kalyuzhnyy N A 2016 Semiconductors 50 517
[14] Rybalchenko D V, Mintairov S A, Shvarts M Z and Kalyuzhnyy N A 2016 Journal of Physics: Conference Series 741 012086
[15] Nadtochiy A M, Mintairov S A, Kalyuzhnyy N A, Rouvimov S S, Sheryakov Y M, Payusov A S, Maximov M V and Zhukov A E 2015 Semiconductors 49 1090-1094