Aalto University

Aaltodoc

OPEN ACCESS

Author(s): Hakkarainen, T. & Toivonen, J. & Sopanen, M. & Lipsanen, Harri

Title: Self-assembled GaIn(N)As quantum dots: Enhanced luminescence at 1.3 μm

Year: 2001

Version: Final published version

Self-assembled GaIn(N)As quantum dots: Enhanced luminescence at 1.3 μm

T. Hakkarainen, J. Toivonen, M. Sopanen, and H. Lipsanen

Self-assembled GaIn(N)As quantum dots: Enhanced luminescence at 1.3 μm

T. Hakkarainen,[a] J. Toivonen, M. Sopanen, and H. Lipsanen

Optoelectronics Laboratory, Helsinki University of Technology, P.O. Box 3000, FIN-02015 HUT, Finland

Self-assembled GaIn(N)As quantum dots are fabricated on GaAs by atmospheric pressure metalorganic vapor-phase epitaxy using dimethylhydrazine (DMHy) precursor as a nitrogen source. The incorporation of nitrogen into the islands is observed to be negligible. However, the areal density of the islands is increased by up to one order of magnitude compared to that of the respective GaInAs islands. The GaIn(N)As island size can also be controlled by varying the DMHy flow. An enhancement of the room-temperature luminescence at 1.3 μm is observed in the GaIn(N)As samples grown with DMHy. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1425082]

In the last few years there has been considerable attention to material and device research for 1.3 and 1.55 μm laser structures grown on GaAs substrates. One method to achieve 1.3 μm laser emission on GaAs is to use self-assembled GaInAs quantum dots (QDs) in the active region.[1,2] The same emission wavelength can also be achieved with InAs QDs embedded in GaAs[3] or in GaInAs.[4,5] The GaInNAs alloy has recently been proposed as a possible material for long wavelength lasers,[6] and GaInNAs/GaAs quantum well (QW) lasers with emission wavelengths around 1.3 μm have been fabricated by molecular beam epitaxy (MBE)[7–9] and by metalorganic vapor-phase epitaxy (MOVPE).[10,11] Solid-source MBE has been used to fabricate GaInNAs/GaAs QW laser diodes operating at 1.52 μm.[12] Room-temperature photoluminescence at 1.52 μm has recently been reported from GaInNAs QDs grown by gas-source MBE.[13] GaInNAs QDs have also been grown by chemical beam epitaxy (CBE),[14] but until now there has been no reports of MOVPE-grown GaInNAs QDs. In this work we have grown self-assembled GaIn(N)As QDs by atmospheric pressure MOVPE and investigated the effect of nitrogen on formation and optical properties of the GaIn(N)As islands.

The samples were grown on semi-insulating GaAs (100) substrates in a horizontal MOVPE reactor at atmospheric pressure. Trimethylgallium (TMGa), trimethylindium (TMIn), tertiarybutylarsine (TBAs), and dimethylhydrazine (DMHy) were used as sources for gallium, indium, arsenic, and nitrogen, respectively. The sample structure consists of a 100-nm-thick GaAs buffer layer grown at 650 °C, a layer of $Ga_{1-x}In_x$(N)As islands with the In composition of $x=0.45$ or $x=0.6$ and a GaAs cap layer in some of the samples. The GaIn(N)As layer and the cap layer were grown at the same temperature, varied in the range of 450–570 °C. All the temperatures mentioned here are thermocouple readings. The nominal growth rate and the nominal thickness were varied in the range of 0.2–4 ML/s and 2.5–8 ML, respectively. The uncovered islands on the sample surface were imaged by atomic force microscopy (AFM) using SiN tips. The optical

properties of the covered islands were investigated by low temperature (LT), i.e., 10 K, and room-temperature (RT) photoluminescence (PL) measurements. An argon ion laser was used for excitation and the luminescence was detected by a liquid-nitrogen-cooled Ge detector.

To investigate how the nominal coverage affects the island formation, uncovered islands with the In composition of $x=0.6$ were grown at 530 °C with the DMHy/TBAs ratio of 16 and the nominal growth rate of 1.3 ML/s. This growth rate was chosen because the island density was observed to be highest within a growth rate range of 1.3–2 ML/s. Figure 1(a) shows an AFM image from a 3 ML $Ga_{0.4}In_{0.6}$(N)As sample with an island density of 3×10^8 cm^{-2} and an average island height of 8 nm. The coverage of 3 ML is close to the threshold thickness for island formation since no islands were observed when the coverage was decreased to 2.5 ML. Figures 1(b) and 1(c) show AFM images from 3.5 and 4.5 ML $Ga_{0.4}In_{0.6}$(N)As samples, respectively. The areal density and the average height of the islands in the 4.5 ML sample are 5×10^{10} cm^{-2} and 5 nm, respectively. An AFM image
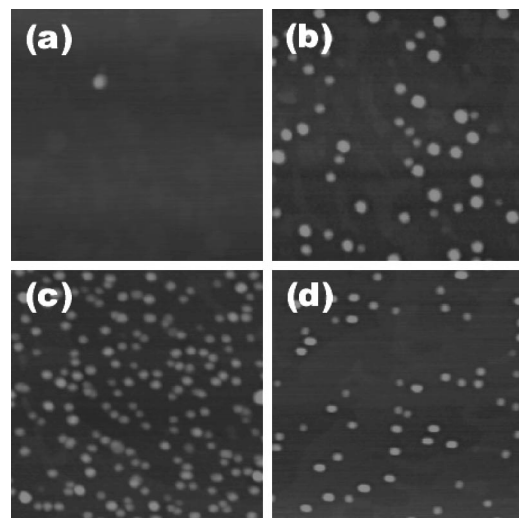

FIG. 1. AFM images taken from QD samples grown at 530 °C with (a) 3 ML $Ga_{0.4}In_{0.6}$(N)As, (b) 3.5 ML $Ga_{0.4}In_{0.6}$(N)As, (c) 4.5 ML $Ga_{0.4}In_{0.6}$(N)As, and (d) 4.5 ML $Ga_{0.4}In_{0.6}$As. The scan size is 0.5 $\times$0.5 μm^2.

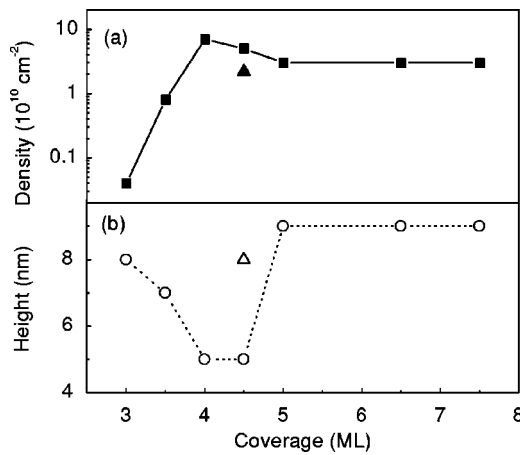[a]Author to whom correspondence should be addressed; electronic mail: thakkara@cc.hut.fi

FIG. 2. (a) Island density and (b) average island height measured by AFM as a function of coverage for $Ga_{0.4}In_{0.6}$(N)As QD samples grown at 530 °C. The triangles show the respective data for the reference $Ga_{0.4}In_{0.6}$As QD sample.

from a 4.5 ML $Ga_{0.4}In_{0.6}$As sample grown without DMHy flow is shown in Fig. 1(d). The island density in this N-free sample is smaller and the islands are higher on average compared to the sample grown with DMHy flow. This is in contrast to the results that were observed for MBE growth.[13] Figures 2(a) and 2(b) show the island density and the average island height, respectively, as a function of coverage. The maximum island density and the minimum island height are achieved with the coverage of 4 ML. When the coverage is increased, the island density remains fairly constant in the range of $3-4 \times 10^{10} \, cm^{-2}$ and the average height of the islands increases to 9 nm. Also larger, relaxed islands with an areal density of $10^7 - 10^9 \, cm^{-2}$ are observed in the samples. The areal density of these relaxed islands increases with increasing coverage.

To investigate the optical properties of the islands, the islands were covered with a 50-nm-thick layer of GaAs. Figure 3 shows LT-PL spectra from three QD samples: (a) 3.5 ML $Ga_{0.4}In_{0.6}$(N)As, (b) 4.5 ML $Ga_{0.4}In_{0.6}$(N)As, and (c) 4.5 ML $Ga_{0.4}In_{0.6}$As. The major peak in each spectrum originates from the covered islands acting as QDs. As the coverage is increased from 3.5 to 4.5 ML the PL intensity decreases since the density of large, incoherent islands increases. The PL peak of the $Ga_{0.4}In_{0.6}$As reference sample is at smaller energy compared to the sample grown with DMHy flow. This indicates that a negligible amount of nitro-
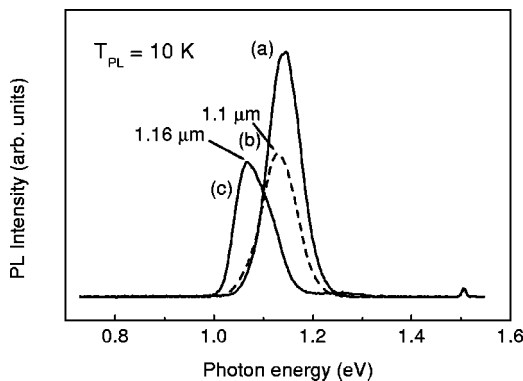

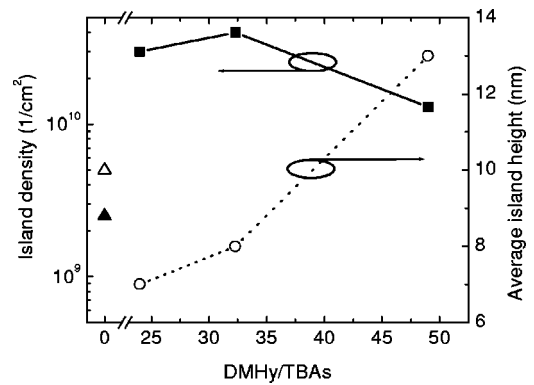FIG. 3. LT-PL spectra from (a) 3.5 ML $Ga_{0.4}In_{0.6}$(N)As, (b) 4.5 ML $Ga_{0.4}In_{0.6}$(N)As, and (c) 4.5 ML $Ga_{0.4}In_{0.6}$As QD samples grown at 530 °C.

FIG. 4. Island density and average island height as a function of DMHy/TBAs ratio for 8 ML $Ga_{0.55}In_{0.45}$(N)As QD samples grown at 520 °C. The triangles show the respective data for the reference $Ga_{0.55}In_{0.45}$As QD sample.

gen is incorporated into the islands. The energy difference is explained by the fact that the average height of the $Ga_{0.4}In_{0.6}$As islands is larger than that of the $Ga_{0.4}In_{0.6}$(N)As islands. The full width at half maximum of the PL peak is about the same for samples (b) and (c). The reason for the negligible incorporation of nitrogen lies behind the large In composition of the islands. It has been observed that the incorporation of nitrogen is seriously hindered by the presence of In during MOVPE growth of GaInNAs.[15] The mechanism behind this behavior is not completely understood yet, although it has been speculated that it could be caused by surface segregation of In.[16] An attempt was made to increase the incorporation of nitrogen by decreasing the growth temperature but this only resulted in degradation of the optical quality.

To study whether more nitrogen could be incorporated into GaInNAs with lower In composition, samples with an In composition of $x = 0.45$ were grown. Since the wetting layer may become thicker due to the decreasing lattice mismatch with decreasing In composition, the nominal coverage was chosen to be 8 ML to make sure that islands are formed. Figure 4 shows the dependence of the island density and the average island height on the DMHy/TBAs ratio. Also shown are the data for a $Ga_{0.55}In_{0.45}$As reference sample. The island densities in the $Ga_{0.55}In_{0.45}$(N)As QD samples are significantly larger compared to the reference sample for all the used DMHy/TBAs ratios. When the DMHy/TBAs ratio is increased from 24 to 49 the average island height increases from 7 to 13 nm. The height of the $Ga_{0.55}In_{0.45}$As islands is 10 nm on average. Thus, by using DMHy during the island growth the island density can be increased by one order of magnitude. On the other hand, the size of the islands can be determined by choosing the appropriate DMHy/TBAs ratio.

Figures 5(a) and 5(b) show the RT-PL spectra from $Ga_{0.55}In_{0.45}$(N)As and $Ga_{0.55}In_{0.45}$As QD samples, respectively. The $Ga_{0.55}In_{0.45}$(N)As sample was grown with a DMHy/TBAs ratio of 24 and it has about one order of magnitude larger island density than the $Ga_{0.55}In_{0.45}$As reference sample. The spectrum of the reference sample exhibits a QD ground state transition at 0.93 eV (1.33 μm) and a dominating wetting layer transition at 1.18 eV (1.05 μm). One can also see an excited state transition 76 meV higher in energy compared to the ground state transition. On the contrary, the
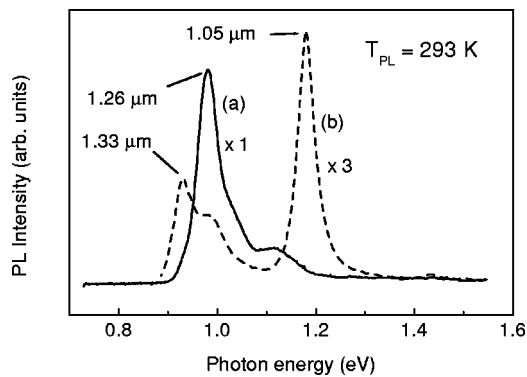
FIG. 5. RT-PL spectra from 8 ML (a) $Ga_{0.55}In_{0.45}$(N)As and (b) $Ga_{0.55}In_{0.45}$As QD samples grown at 520 °C.

$Ga_{0.55}In_{0.45}$(N)As sample shows no wetting layer transition indicating efficient capture of the carriers into the QDs. One can see both a well pronounced ground state transition at 0.98 eV (1.26 μm) and an excited state transition 130 meV higher in energy. The integrated PL intensity from the $Ga_{0.55}In_{0.45}$(N)As sample is about 3 times larger than from the $Ga_{0.55}In_{0.45}$As QDs in Fig. 5(b). The difference in the ground state peak emission wavelength between the two samples can again be explained by the larger size of the $Ga_{0.55}In_{0.45}$As QDs. LT-PL measurements of the $Ga_{0.55}In_{0.45}$(N)As samples grown with different DMHy/TBAs ratios are consistent with the assumption that the incorporation of nitrogen is negligible. This shows that the incorporation of nitrogen into GaInNAs QDs grown by atmospheric pressure MOVPE cannot be increased simply by increasing the DMHy/TBAs ratio. However, the use of DMHy flow during the growth of GaIn(N)As islands improves the areal density of the islands and enhances the PL intensity in the covered samples.

In summary, self-assembled GaIn(N)As QDs were grown by atmospheric pressure MOVPE. The areal density of the GaIn(N)As islands is higher than that of the GaInAs islands grown with similar parameters. The island height of the GaIn(N)As islands can be varied to some extent by changing the DMHy/TBAs ratio. PL studies of overgrown islands indicate that the incorporation of nitrogen is negligible. However, an enhancement of the 1.3 μm RT luminescence is observed. Thus, by introducing DMHy during the island growth the optical properties of the GaIn(N)As islands can be improved.

[1] D. L. Huffaker, G. Park, Z. Zou, O. B. Shchekin, and D. G. Deppe, Appl. Phys. Lett. **73**, 2564 (1998).
[2] G. Park, O. B. Shchekin, S. Csutak, D. L. Huffaker, and D. G. Deppe, Appl. Phys. Lett. **75**, 3267 (1999).
[3] I. Mukhametzhanov, R. Heitz, J. Zeng, P. Chen, and A. Madhukar, Appl. Phys. Lett. **73**, 1841 (1998).
[4] V. M. Ustinov, N. A. Maleev, A. E. Zhukov, A. R. Kovsh, A. Yu. Egorov, A. V. Lunev, B. V. Volovik, I. L. Krestnikov, Yu. G. Musikhin, N. A. Bert, P. S. Kop'ev, Zh. I. Alferov, N. N. Ledentsov, and D. Bimberg, Appl. Phys. Lett. **74**, 2815 (1999).
[5] K. Nishi, H. Saito, S. Sugou, and J.-S. Lee, Appl. Phys. Lett. **74**, 1111 (1999).
[6] M. Kondow, K. Uomi, A. Niwa, T. Kitatani, S. Watahiki, and Y. Yazawa, Jpn. J. Appl. Phys., Part 1 **35**, 1273 (1996).
[7] K. Nakahara, M. Kondow, T. Kitatani, M. Larson, and K. Uomi, IEEE Photonics Technol. Lett. **10**, 487 (1998).
[8] A. Yu. Egorov, D. Bernklau, D. Livshits, V. Ustinov, Zh. I. Alferov, and H. Riechert, Electron. Lett. **35**, 1643 (1999).
[9] M. Reinhardt, M. Fischer, M. Kamp, J. Hofmann, and A. Forchel, IEEE Photonics Technol. Lett. **12**, 239 (2000).
[10] F. Höhnsdorf, J. Koch, S. Leu, W. Stolz, B. Borchert, and M. Druminski, Electron. Lett. **35**, 571 (1999).
[11] C. Ellmers, F. Höhnsdorf, J. Koch, C. Agert, S. Leu, D. Karaiskaj, M. Hofmann, W. Stolz, and W. W. Rühle, Appl. Phys. Lett. **74**, 2271 (1999).
[12] M. Fischer, M. Reinhardt, and A. Forchel, Electron. Lett. **36**, 1208 (2000).
[13] M. Sopanen, H. P. Xin, and C. W. Tu, Appl. Phys. Lett. **76**, 994 (2000).
[14] S. Makino, T. Miyamoto, K. Nageyama, F. Koyama, and K. Iga, J. Cryst. Growth **221**, 561 (2000).
[15] R. Bhat, C. Caneau, L. Salamanca-Riba, W. Bi, and C. Tu, J. Cryst. Growth **195**, 427 (1998).
[16] D. J. Friedman, J. F. Geisz, S. R. Kurtz, J. M. Olson, and R. Reedy, J. Cryst. Growth **195**, 438 (1998).