



PROMIS – Postgraduate Research on Dilute Metamorphic Nanostructures and Metamaterials in Semiconductor Photonics

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Report on Deliverable D 1.3: Optimal hydrogenation conditions for full HN passivation determined

Introduction

Recently, a novel route for the fabrication of site-controlled nanostructures based on hydrogen-assisted defect engineering in dilute nitride semiconductors has been demonstrated [1]. This novel and original fabrication method combines the precision of modern electron-beam-lithography systems—which allow for the deposition of ordered arrays of nanometer-sized metallic structures on the surface of a sample—with the striking capability of hydrogen to modulate the electronic, optical, electric, and structural properties of dilute nitrides, such as GaAsN [2]. H-induced modulation effects on the physical properties of GaAsN is due to the formation of stable N-2H-H complexes [3], which gradually and reversibly *transmute* dilute nitrides in (effectively) N-free alloys by neutralizing the effects of nitrogen on the properties of the host crystal. By spatially controlling the hydrogen-nitrogen interaction at the nanometer scale (and consequently the band-gap energy in all spatial directions), site-controlled nanostructures with optical spectra resembling those of self-assembled semiconductor QDs have been fabricated [4].

Research objectives and background

So far, much effort has been posed on GaAsN, since this material offers excellent light sources without exhibiting strong line broadening effects. However, in order to attain emission wavelength close to 1.3 and 1.55 μm , InGaAsN is preferable thanks to the simultaneous red-shift of the band gap energy caused by both N and In incorporation. Furthermore, N tends to counteract the compressive strain present in InGaAs/GaAs heterostructures that can possibly lead to a perfect lattice matching of InGaAsN on GaAs substrates.

Despite the first observation of N passivation in dilute nitrides was reported in InGaAsN quantum wells [5], not much work was done afterwards. In the following, we report on a detailed study of hydrogenation conditions on different InGaAsN/GaAs heterostructures grown under different conditions and by different MOVPE laboratories.

Description of Work

Testing of hydrogen effects on the electronic properties of InGaAsN quantum wells (QWs) was carried out on samples provided by the Philips University at Marburg (UMR). Photoluminescence (PL) measurements have been employed to assess the photon emission spectra of the InGaAsN QWs before and after H irradiation. The untreated samples, grown using MOVPE at UMR, consisted of a InGaAsN/GaAs/InGaAsN structure sandwiched between two GaAs buffer and capping layers, all grown on a GaAs substrate (Fig. 1). Both InGaAsN layers were grown to be identical and 7 nm thick, with nominal Indium concentration equal to 28% and N concentration equal to 2.8%. Four different samples were studied. The samples differ in the growth rate (slow: 0.045nm/s vs fast: 0.24nm/s) and Ga precursor employed (triethyl-Ga, TEGa, vs trimethyl-Ga, TMGa).

138 nm GaAs	sample	Growth rate	Ga precursor
7 nm InGaAsN	14464	Slow	TEGa
20 nm GaAs	14465	Fast	TEGa
7 nm InGaAsN	14469	Slow	TMGa
138 nm GaAs	14470	Fast	TMGa

Figure 1. The sample layer composition. Notice that there are two InGaAsN QWs. The structure was grown by MOVPE on a GaAs substrate by using different growth conditions.

Results

Figure 2 shows the PL spectra at room temperature of the untreated samples as received from UMR. The spectral peaks are around 1 and 1.05 eV. One can notice a 3-4 times lower emission intensity for sample 14464 compared to the other samples.

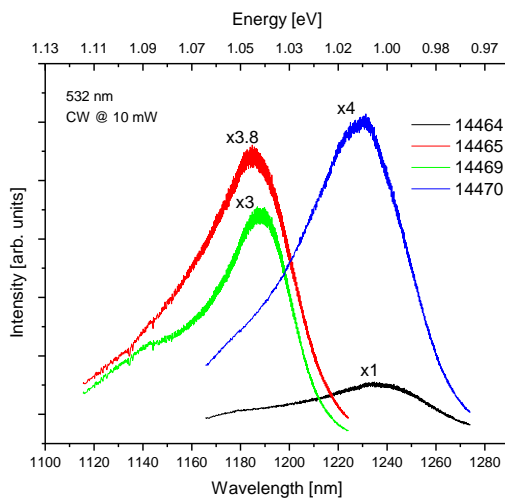


Figure 2. Photoluminescence (PL) spectra recorded at room temperature of samples 144xx. Notice the very low emission intensity of sample 14464 relative to the other samples.

In terms of spatial quality of the QW layers, as the laser excitation spot is much smaller than the sample size, we were able to perform peak mapping of each of the samples and found that the peak wavelength changes, with respect to the excitation spot position, by $\Delta\lambda$ ranging between ~20-50 nm, as shown in Figure 3.

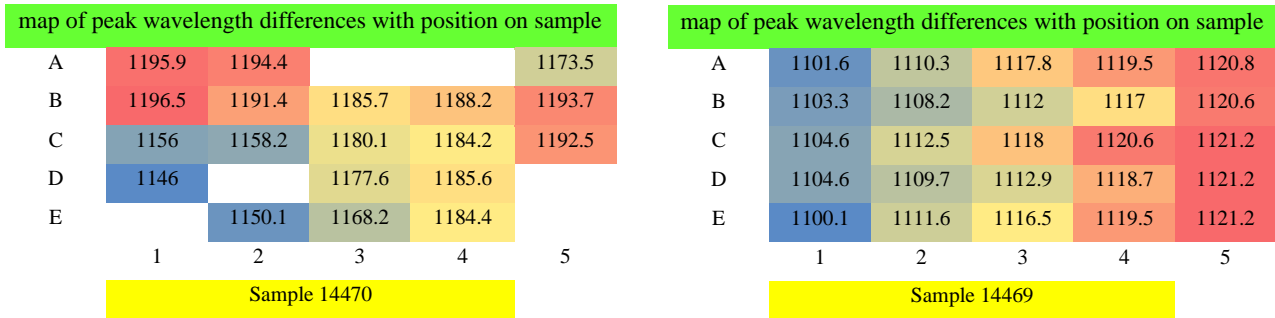


Figure 3. Photoluminescence (PL) spectra peak intensities as a function to excitation position across the sample surface. Samples were roughly $5 \times 5 \text{ mm}^2$ so the difference between adjacent points is about 1mm. Spectra were recorded at 10K with excitation power of 0.1 mW.

The outcome of sample treatment with H doses equal to 5×10^{16} (5A) and 2×10^{17} (2B) ions/cm² at 190 °C is displayed in Figures 4a and 4b. The PL spectra were recorded at $T=10 \text{ K}$ because H was found to cause a considerable decrease in the PL signal at RT. The origin of this large decrease in the emission efficiency is not yet understood. It is assumed that, in the case of InGaAsN, the presence of In leads likely to an augmented “fragility” of the crystal with respect to defects induced by the hydrogenation process. This effect needs further investigation and it will be studied further. Lowering the temperature allowed also the use of a much lower excitation power, thus reducing line shape broadening effects. The most important observation in these spectra, was the lack of any sizable blue-shift of the PL peaks following hydrogenation, contrary to what was expected. In particular, after introducing the much higher H dose of 2×10^{17} ions/cm² the intensity ratios between the untreated and the hydrogenated samples were not observed to be different than when using the lower dose. The initial assumption as to the cause of these results was that the temperature used in these hydrogenations, 190 °C, was too low to allow sufficient diffusion of hydrogen into the QWs. To verify this, samples that were hydrogenated previously with 5×10^{16} dose at 190 °C, were hydrogenated again with the same dose but at 250 °C. The results of this attempt, shown in Figure 4c, don’t support our assumption as intensity ratios between untreated and hydrogenated samples remained the same and no pronounced blue-shifting was observed either. At this point, serious doubts as to the integrity of our hydrogenation apparatus, started to creep in and to rule this out, we used previously studied samples fabricated both at UMR (K12882, designated hereafter as UMR-O [6]) and the EPFL (M3920, designated hereafter as EPFL-O) and hydrogenated them together with ours. Both samples were grown by MOVPE. These samples were chosen because of their resemblance to our samples in terms of the layer types fabricated and their dimensions (details can be found in the Appendix). Hydrogenation conditions, namely a H dose of 10^{17} ions/cm² (1B) at 300 °C, were chosen as to simulate previous hydrogenations done successfully on both UMR-O and EPFL-O. The results are displayed in Figure 5, where new peaks, significantly blue-shifted at about 75 and 125

meV, are observed for UMR-O and EPFL-O samples, respectively (inset A), while for our samples, only a significant decrease in the PL intensity can be seen without any pronounced peak-shifting (inset B). These results practically rule out any doubts related to our experimental setup and prove its integrity.

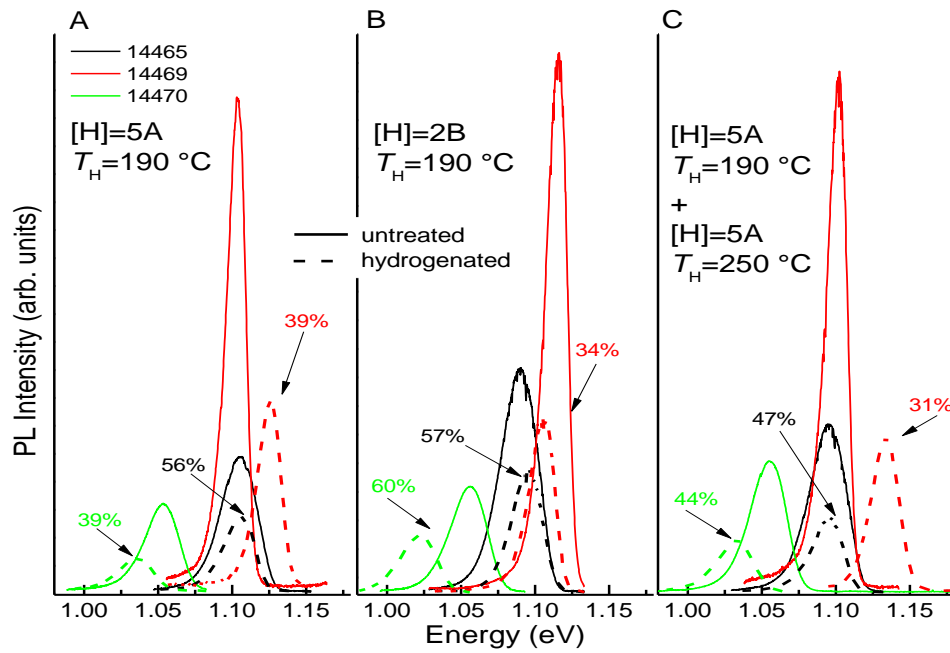


Figure 4. Photoluminescence (PL) spectra of samples 144xx prior and after hydrogenation. Solid lines indicate untreated samples while dashed lines indicate hydrogenated ones. The hydrogenation conditions are found within the insets (detailed information can be found in the text). Spectra were recorded at 10 K with excitation power of 0.1 mW. Numbers pointing at the peaks of hydrogenated samples indicate the relative intensity of each sample compared with its untreated counterpart. The spectra show that while H irradiation has a general effect of lowering the emission intensity, neither the intensity nor the peak energy seem to be affected by the specifics of the hydrogenation; increasing the H dose 4-times did not affect the intensity ratios (insets A and B) so as hydrogenation at 250 °C (inset C).

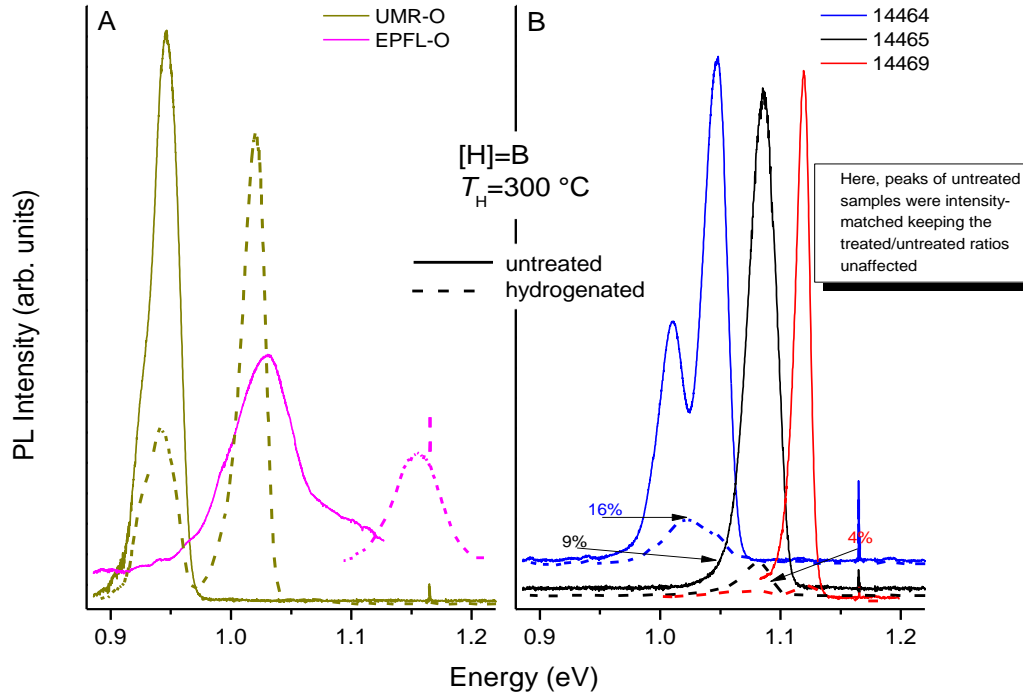


Figure 5. PL spectra of samples UMR-O, EPFL-O and 144xx, prior and after hydrogenation. Solid lines indicate untreated samples while dashed lines indicate hydrogenated ones. The hydrogenation conditions are found within the box inset. Spectra were recorded at 10 K with excitation power of 5 mW for xx-O samples and 0.1 mW for 144xx samples. Numbers pointing at the peaks of hydrogenated samples, in inset B, indicate the relative intensity of each sample compared with its untreated counterpart. The spectra show that while H irradiation successfully (partially) passivated the old samples (inset A), the same irradiation conditions only reduced the original peaks intensities (inset B).

Conclusions

Upon H irradiation of the samples 144xx, recently supplied by UMR, we observe only a reduction in the original peak intensities without any blue-shifting. This reduction is similar for 190 and 250 °C, regardless of the hydrogen dose used, and increases dramatically for hydrogenation performed at 300 °C. These observations, together with the fact that old samples, similar in layer composition and dimensions, were hydrogenated successfully under the same conditions, strongly suggest that samples 144xx are far more susceptible to H-beam/temperature damage than samples fabricated previously. Furthermore, as the two QWs of sample 14464 were found to be slightly different in their PL peak energies, this allowed us to observe that one QW seemed to have improved PL intensity in the hydrogenated case relative to the untreated counterpart while the other was completely erased, see Figure 6. This effect appeared to be gradually and progressively wiped out with increasing H dose and hydrogenation temperature. All these findings, in addition to observing significant inhomogeneity of the central PL peak energy with respect to position across the sample, lead us to recommend rethinking about the best fabrication route for achieving more resilient samples.

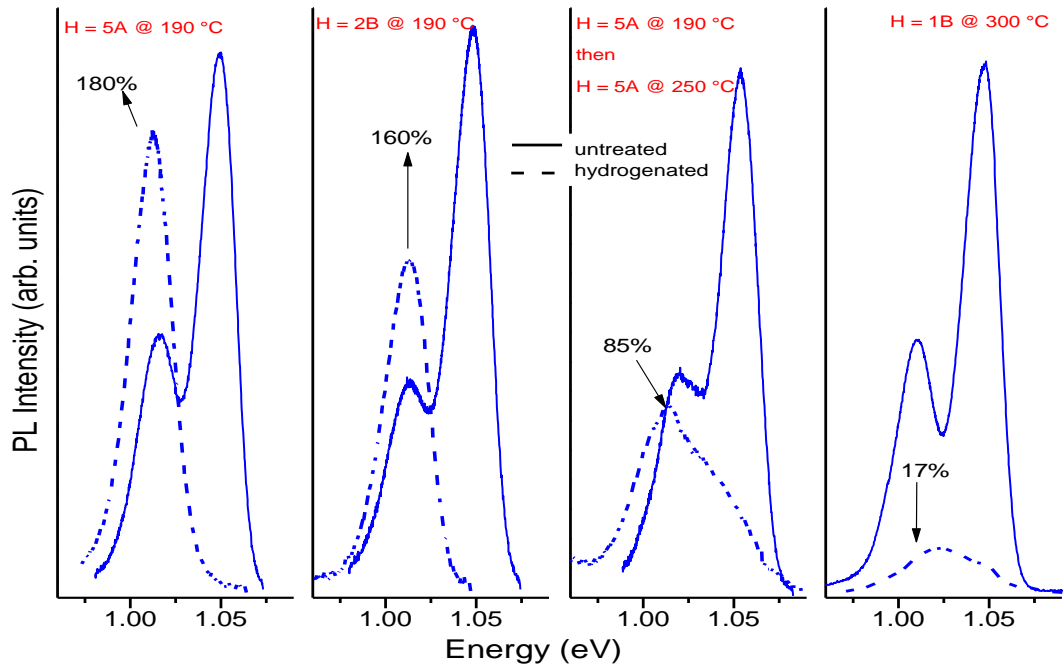


Figure 6. PL spectra from different specimens of sample 14464, prior and after hydrogenation. Solid lines indicate untreated samples while dashed lines indicate hydrogenated ones. The hydrogenation conditions can be seen in red. Spectra were recorded at 10K with excitation power of 0.1mW. Numbers pointing at the peaks of hydrogenated samples, indicate the relative intensity of each hydrogenated sample compared with the intensity of the lower peak of the untreated counterpart.

APPENDIX

We report here the main parameters of UMR-O and EPFL-O samples used as a reference.

UMR-O: 5 quantum wells with thickness equal to 8.3 nm. The QWs are separated by a 58.9 nm-thick GaAs barrier. The composition of the InGaAsN QWs is [In]=30% and [N]=0.6%.

EPFL-O: 2 quantum wells with thickness equal to 5 nm. The QWs are separated by a 50nm-thick GaAs barrier. The composition of the firstly and secondly grown QW is [In]=27% and [N]=1.7% and . [In]=27% and [N]=0%, respectively.

References

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