



Research paper

Influence of GaInNAs/GaAs QWs composition profile on the transitions selection rules



Damian Pucicki *, Katarzyna Bielak, Mikołaj Badura, Wojciech Dawidowski, Beata Ściana

Faculty of Microsystem Electronics and Photonics, Wrocław University of Technology, Z. Janiszewskiego 11/17, 50-372, Wrocław, Poland

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ABSTRACT

In this work authors present the results of structural and optical characterization of the GaInNAs/GaAs MQW structures grown by AP-MOVPE. The growth conditions of quantum well layers were modified in order to counteract the phase segregation within GaInNAs QWs. Profiles of indium and nitrogen contents across the QWs layers were investigated by using HRXRD. The optical properties were investigated by applying photoluminescence spectroscopy. The inhomogeneous composition of the GaInNAs QW layers is considered as a factor suppressing the PL intensity due to change in the electron and holes wave functions distributions and tuning the transition's selection rules.

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1. Introduction

Significant progress in the development of multicomponent semiconductors like GaInNAs(Sb) allows extension of the working wavelength range of optoelectronic device structures based on GaAs substrates. Optimization of growth conditions and better understanding of the special features of the investigated material system have resulted in huge progress during the last years. There are several successful methods leading to improvement of the GaInNAs crystal quality as well as reduction of the alloy band gap such as: the use of GaAsN barriers for strain compensation and wavelength extension [1], optimizing the growth temperature and corresponding III/V ratios [2,3], adding Sb to the QW resulting in the better quality of grown material [4–6]. However, regardless of the technique of epitaxial growth of GaInNAs, strong phase segregation occurs in this kind of material [7–10]. The inhomogeneous distributions of indium and nitrogen atoms along the GaInNAs quantum wells (QWs) thicknesses lead to built-in potential in the QW. In agreement with quantum-confined Stark effect (QCSE) this potential affects the emitted wavelength [11] and influences electrons and holes distributions in the well, decreasing the overlap integral of the wave functions. Furthermore, according to the Fermi's golden rule, separation of electrons and holes wave functions changes the transitions selection rules and has influence on the properties of the QW system. In this work we present the improvement of the optical quality of the GaInNAs/GaAs MQW structures by modification of the GaInNAs QW growth. The growth modification was introduced in

order to change the QWs potential distribution and to counteract the electrons and holes spatial segregation within the QW layers.

2. Experimental details

In this work GaInNAs/GaAs MQW structures were investigated. All considered structures were grown by atmospheric pressure metalorganic vapor phase epitaxy (AP-MOVPE) in AIX200 R&D AIXTRON horizontal reactor. The studied structures consist of 450 nm thick GaAs buffer and $3 \times$ GaInNAs/GaAs QWs region capped by $40 \div 50$ nm thick GaAs. (100)-oriented semi-insulating (SI) GaAs wafers were used as substrates. The schematic diagram of the layer stack of investigated structures is presented in Fig. 1. Trimethylgallium (TMGa), trimethylindium (TMIn), tertiarybutylhydrazine (TBHy) and arsine (AsH_3 : 10% mixture in H_2) were used as the growth precursors.

All structures were grown in the same conditions apart from the trimethylindium dosage over the time of QWs growth. First sample #NI107 was grown with the constant flow rate of carrier gas through the saturators with the precursors, during growth of GaInNAs QW layers. Referring to previous authors' work [7,8] and what is reported in the literature [9, 10] the phase segregation should occur in this sample. The growth procedures of next two GaInNAs/GaAs MQW structures (#NI114 and #NI118) were modified in order to change the In and N distribution along the GaInNAs quantum wells. The partial pressure of TMIn in the reactor has been changed over the time of QWs growth (90 s). Despite this change, total amount of trimethylindium delivered to the reactor was kept constant, to be the same as in the case of quantum wells of #NI107 structure. In the case of the #NI118 structure the greater change of the TMIn partial pressure over QW growth time was applied. The crucial growth parameters (T_g -

* Corresponding author.

E-mail address: Damian.Pucicki@pwr.edu.pl (D. Pucicki).

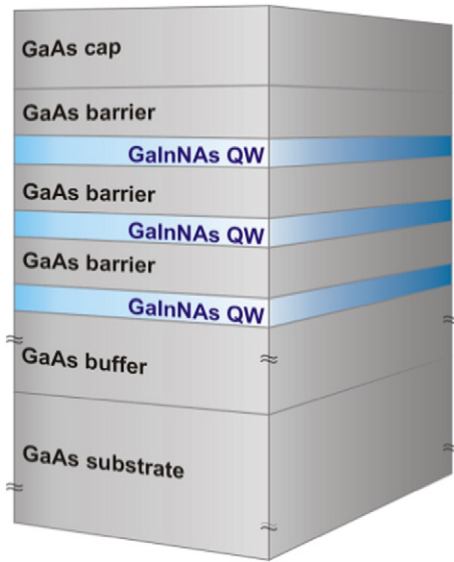


Fig. 1. Illustration of layer stack of the $3 \times$ GaInNAs/GaAs MQW structures.

temperature of growth, p_{TMGa} , p_{TMIIn} , p_{TBHy} , and p_{AsH_3} – partial pressures of used precursors TMGa, TMIIn, TBHy and AsH₃ respectively and $V_{\text{H}_2/\text{TMIIn}}$ – the flow rate of the hydrogen through the TMIIn saturator) of the discussed structures are collected in Table 1.

The structural properties of investigated GaInNAs/GaAs MQW samples were determined by means of high resolution X-ray diffractometry (HRXRD). Unfortunately, in the case of quaternary GaInNAs alloys it is impossible to evaluate simultaneously the indium and nitrogen concentrations from the HRXRD analysis. For that purpose, authors of the work developed the new algorithm of structural characterization of the semiconductor structures which contains GaInNAs QWs. The mentioned algorithm is based on the HRXRD measurements analysis supported by contactless electro-reflectance (CER) or photoreflectance (PR) modulated spectroscopy measurements and by the indium and nitrogen contents calculations based on the band anticrossing (BAC) theory as well. The detailed description of the applied algorithm for structural characterization of investigated structures can be found elsewhere [8]. The measured X-ray diffraction curves of the (004) symmetrical reflexes, of the three discussed samples, were analyzed using dynamical diffraction theory and in agreement with algorithm elaborated by the authors. X'Pert Epitaxy software of PANalytical B.V. was used for diffractograms simulations. That software allows analysis of properties of heteroepitaxial structures such as compositions, thicknesses, mismatch and percentage of relaxation of following layers [12]. The exact data of examined samples were achieved by fitting the simulated curves to the measured one. The thicknesses of following layers of the MQW structures as well as the compositions and inhomogeneities of GaInNAs QWs were the parameters of fitting procedures.

The optical properties of MQW structures were investigated by means of room temperature (RT) photoluminescence (PL) before and after annealing. It was necessary to anneal the investigated samples because of strong suppression of the PL intensity in as-grown structures.

Table 1
Parameters of growth of the GaInNAs QWs structures.

Structure no.	p_{TMGa} (mbar)	$V_{\text{H}_2/\text{TMIIn}}$ (ml/min)	p_{TMIIn} (mbar)	p_{TBHy} (mbar)	p_{AsH_3} (mbar)	T_g (°C)
#NI107	0.03326	35	0.006933	1.447	0.5201	585
#NI114	0.03331 to 0.3321	50–20	0.009919 to 0.003955	1.449 to 1.444	0.5209 to 0.5193	585
#NI118	0.03335 to 0.03317	60–10	0.01192 to 0.001976	1.45 to 1.443	0.5214 to 0.5187	585

Annealing was carried out in order to improve the optical performance of the GaInNAs layers, what is also often reported in the literature [13]. The analyzed samples exhibit low optical quality due to atmospheric pressure conditions of the epitaxial growth, what leads to high defect concentrations, especially in the case of dilute nitride materials. The improvement of the optical quality of the dilute nitrides materials is a result of the thermally activated atomic bonds rearrangement and reduction of the nitrogen related structural defects, which were formed during the growth [14]. Although, the thermal annealing (TA) carried out in nitrogen atmosphere at 700 °C were considered, we have decided to perform more sophisticated method of rapid optical annealing (ROA). To properly compare the optical properties of the investigated samples the annealing process has to be performed carefully in order to prevent overannealing of the samples [13]. The precise control of the annealing process is possible only in the case of the rapid optical annealing. This method provides the opportunity to observe the evolution of PL intensity during the annealing and to terminate the process at optimal conditions, before the structures will be overannealed. For this reasons, for the purpose of this study only the results achieved after ROA were used.

As a results of HRXRD analysis supported by modulated spectroscopy measurements and calculations of the energy structure, the final GaInNAs/GaAs quantum wells electronic structures were achieved. The calculations of QWs energy band structure were performed: within the framework of the usual envelope function approximation, on the basis of the Pikus–Bir Hamiltonian calculations and including hydrostatic and shear stresses. The single band formalism was applied for valence band calculations in order to avoid a time-consuming calculations repeated simultaneously together with the analysis of HRXRD curves. The numerical matrix methods were employed in order to solve Schrödinger equation and to calculate eigenvalues and eigenvectors. More details of utilized band structures calculation method can be found elsewhere [8].

3. Results and discussion

The previous authors' investigation of the AP-MOVPE growth of GaInNAs/GaAs MQW structures provided information about the delay of nitrogen incorporation occurred at the beginning of QWs growth, at applied growth conditions. That effect was taken into account in case of investigation of structural parameters of discussed structures. That assumption is in a good agreement with the HRXRD measurements, what has been analyzed in accordance with the algorithm of structural characterization elaborated by the authors [8]. Such inhomogeneous composition of GaInNAs quantum wells results in a spatial change of the QWs potential.

In Fig. 2 the (004) $\omega/2\theta$ diffraction curves of discussed GaInNAs/GaAs MQW structures are presented, together with the results of fitting. The structural parameters of the GaInNAs QWs, determined from the HRXRD curves analysis, are collected in Table 2. There are specified

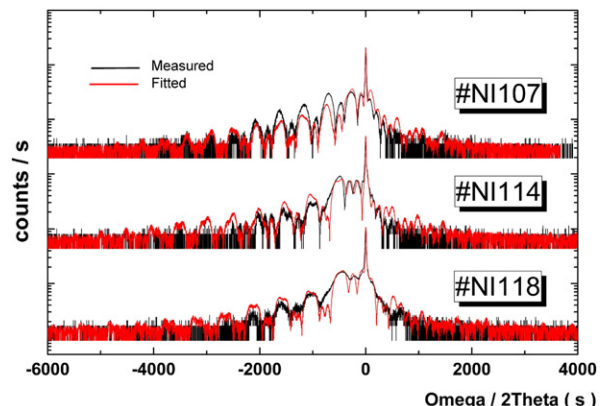


Fig. 2. The (004) $\omega/2\theta$ diffraction curves of GaInNAs/GaAs MQW structures.

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