TECHNOLOGY AND CHARACTERIZATION OF AIIIBV-N HETEROSTRUCTURES FOR SOLAR CELLS APPLICATIONS

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1. Introduction

Unusual properties of GaAsN and InGaAsN semiconductor alloys, so called diluted nitrides - AIIIBV-N, are very attractive for 1.3-1.55 μm lasers [1] and very efficient multijunction solar cells applications [2]. This work presents the influence of the growth parameters such as growth temperature and the gas phase composition on the material quality and nitrogen content in the triple quantum wells 3×InGaAsN/GaAs obtained by atmospheric pressure metal organic vapour phase epitaxy AP-MOVPE. The structural and optical properties of the obtained heterostructures were examined using high resolution X-Ray diffraction (HRXRD), contactless electro-reflectance spectroscopy CER, photoluminescence PL, secondary ion mass spectrometry (SIMS) and transmission electron microscopy (TEM).

2. Experimental details

The InGaAsN/GaAs heterostructures were grown on Si-doped n-type GaAs and semi-insulating SI GaAs substrates by AP-MOVPE using AIX200 R&D Aixtron reactor. Trimethylgallium (TMGa), trimethylindium (TMIn), tertiarybutylhydrazine (TBHy) and 10 % mixture of AsH_3 in H_2 were used as growth precursors, while a high purity H_2 was used as a carrier gas. Undoped multiple quantum wells (MQW) structures consisted of 450 nm thick GaAs buffer and $3 \times In_yGa_{1-y}As_{1-x}N_x$/GaAs triple quantum wells region capped by $40 \div 50$ nm thick GaAs. In order to optimize the epitaxial process the following parameters were varied: growth temperature T_g in range of 566 to 585 °C and the hydrogen flow through the saturator with the organic nitrogen source ($V_{H2/TBHy}$ parameter) from 1100 to 3000 ml/min. Stable parameters were as follows: the low arsine flow rate of 50 ml/ min for InGaAsN quantum wells to increase the nitrogen incorporation and higher arsine flow of 300 ml/ min in the case of GaAs barrier; the ratio of the gallium to indium source concentration in the gas phase $III_{Ga}/III_{In} = 4.8$.

Structural properties of the investigated heterostructures were analysed by measurements of diffraction curves using high resolution X-Ray diffraction (HRXRD). Simulations of the obtained curves allowed the determination of the thickness and composition of the MQW region subject to additional information about the electronic structure of the InGaAsN quantum wells. So, the measurements using contactless electro-reflectance modulation spectroscopy (CER) and photoluminescence (PL) spectroscopy were carried out. The ground state energy of the InGaAsN quantum wells was determined from CER and PL spectra. The quality and uniformity of the MQW structures were verified by SIMS and TEM investigations.

3. Results and discussion

The epitaxial growth of MQW InGaAsN/GaAs structures requires higher growth temperatures (T_g) and lower values of the nitrogen source ($V_{H2/TBHy}$) for good structural and optical quality. The optimal growth parameters are as follows: T_g=585 °C, $V_{H2/TBHy}$=1500 ml/min. Nitrogen incorporation into the InGaAsN quantum wells is less efficient than in the case of GaAsN epilayers grown at the same process conditions what is connected with the weaker In-N bond strength.

Structural properties of the obtained MQW heterostructures were analysed by measurements of diffraction curves of the (004) reflection in the ω/2θ geometry what is shown in Fig. 1. The peak with the highest intensity comes from the GaAs substrate while the satellite peaks are from the MQW structure. Sharpness of these peaks indicates the good crystalline quality and the abruptness of the interfaces. Presence of the satellite oscillations allowed determination of the composition and thickness of InGaAsN quantum wells (16 nm) and GaAs barriers (23 nm) by comparison the measured and simulated diffraction curves. In the case of InGaAsN wells the performed simulations showed that the composition is non-homogeneous, probably due to an interdiffusion of indium into GaAs barrier [3]. The higher indium concentration of about 16% is located inside 9 nm thick InGaAsN QW at the surface side interface while in the next part of 7 nm thick QW the indium content drops to 8%. The nitrogen concentration slightly decreases from 0.18 % to 0.2%, respectively.

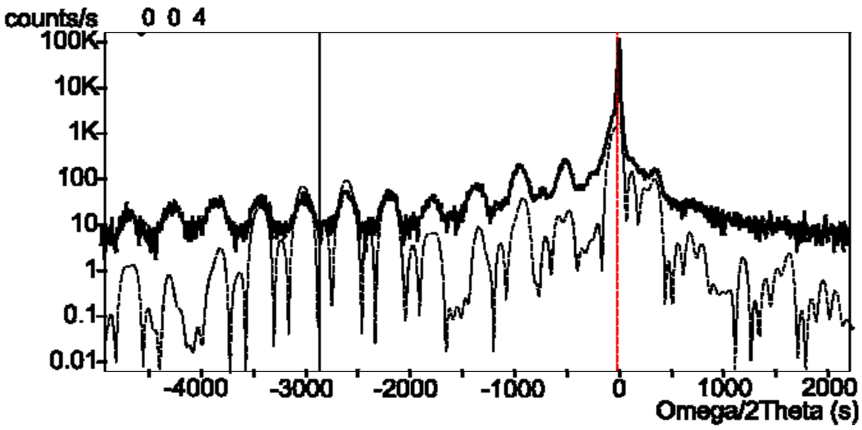

Fig.1: *Measured (solid line) and simulated (dash line) diffraction curves of (004) reflection of InGaAsN/GaAs MQW heterostructure (sample NI 46n).*

TEM image in bright field of InGaAsN/GaAs MQW structure is shown in Fig. 2a. The interfaces between InGaAsN quantum wells and GaAs barriers are very sharp. Different contrast inside quantum wells indicates that they consist of two homogenous layers with the thickness of 6 nm and 10 nm, the barrier thickness between QW is 26 nm. The obtained results confirm non-uniform indium content inside InGaAsN quantum well what corresponds with HRXRD measurements. The electron diffraction image of the mentioned structure is presented in Fig. 2b. Electron diffraction on layer is the same as in substrate, what indicates that QWs are good lattice-matched to GaAs substrate.

a) b)

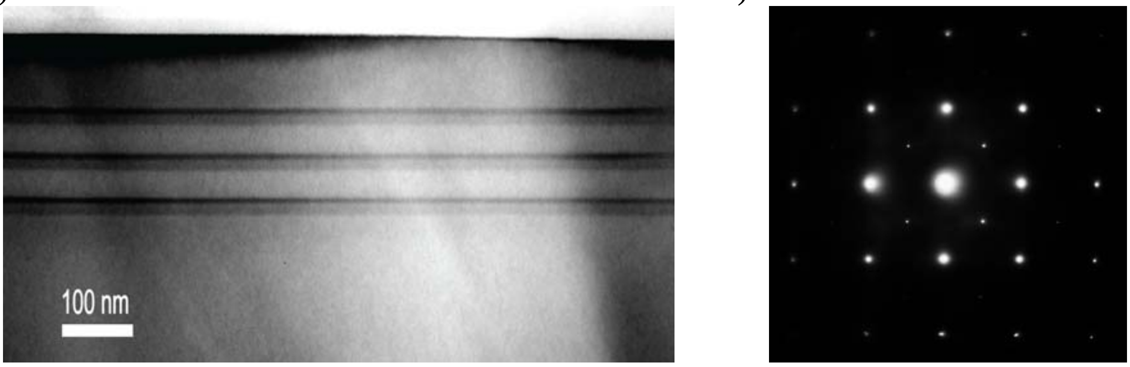

Fig.2: *TEM (a) and electron diffraction (b) images of InGaAsN/GaAs MQW heterostructure (sample NI65n).*

Secondary ion mass spectrometry (SIMS) was applied to get information about the composition profile inside the MQW structures. Fig. 3 shows the results of In and GaN distribution along the growth direction. The presented SIMS profile clearly reveals that indium position in QWs is shifted toward the surface of the sample what can be connected with the interdiffusion process at the InGaAsN/GaAs interface.

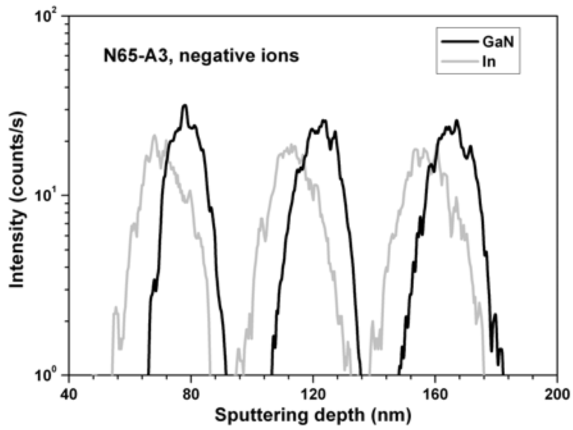

Fig.3: *Composition profile of GaN and In inside the InGaAsN/GaAs MQW region measured by SIMS (sample NI65n).*

Fig. 4a presents PL spectra of the investigated MQW structure measured at 300 K under different annealing conditions: as-grown and after rapid thermal annealing (RTA) at 700 °C for 1 and 5 min. Post-grown annealing distinctly improves the material quality. The intensity of the annealed spectra increases and their broadening decreases. Additionally well-known the blueshift is visible. These effects are connected with a reduction of the hydrogen-related defects in InGaAsN QW, which play the role of nonradiative recombination centres [4]. The best annealing time is 1 min.

CER spectrum of the best MQW structure grown at 585 °C with the hydrogen flow through the saturator with the organic nitrogen source of 1500 ml/min is shown in Fig. 4b. The sample was not annealed. The strong and distinct quantum well transitions appear below the band gap of GaAs (1.43 eV) indicate a good optical quality. Based on QW transitions the energy of the ground state (GS) was determined. This value is about 1.18 eV and is in a good agreement with the result obtained from as-grown PL spectrum (Fig. 4a).

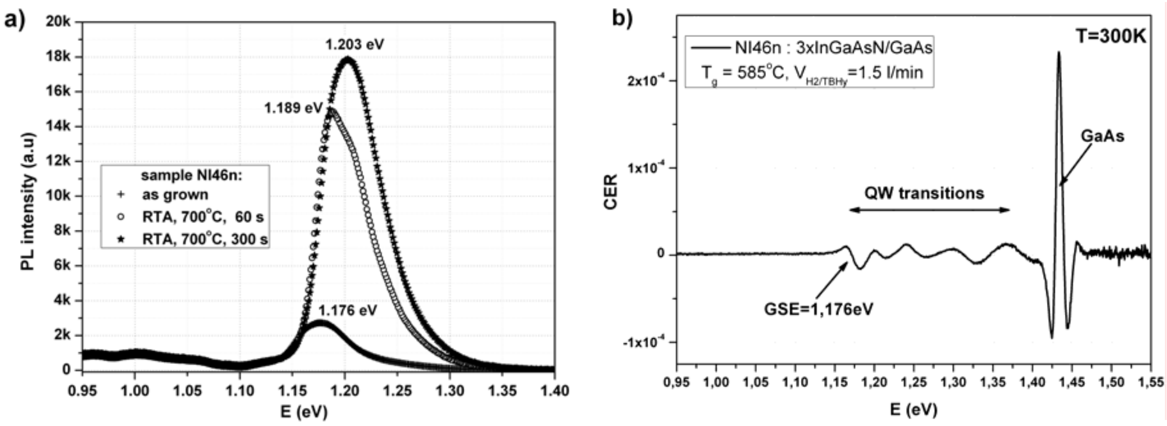

Fig.4: *PL (a) and CER (b) spectra of the InGaAsN/GaAs MQW (sample NI46n).*

4. Conclusions

This work presents the influence of the growth parameters on the structural and optical properties of triple quantum wells 3×InGaAsN/GaAs obtained by atmospheric pressure metal organic vapour phase epitaxy. The MQW InGaAsN/GaAs structures require higher growth temperatures for good structural and optical quality. Higher values of the nitrogen source in the gas phase worsen their optical quality without deterioration of the structural properties. HRXRD, SIMS and TEM investigations showed that the composition inside QWs is non-homogeneous, the maximum of indium concentration shifts toward the surface of the samples. Post-grown annealing distinctly improves the material quality of the InGaAsN/GaAs MQW structures and reveals the well-known effect of the blueshift. Future experimental work will be focused on the understanding and elimination the composition inhomogeneity inside InGaAsN quantum wells. The best MQW structure was introduced into an intrinsic region of the test p-i-n solar cell construction. The measured dc I-U characteristics indicated photoresponse of the cell under the optical excitation of 980 nm what confirms the utility of the InGaAsN material in photovoltaic applications [5].

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