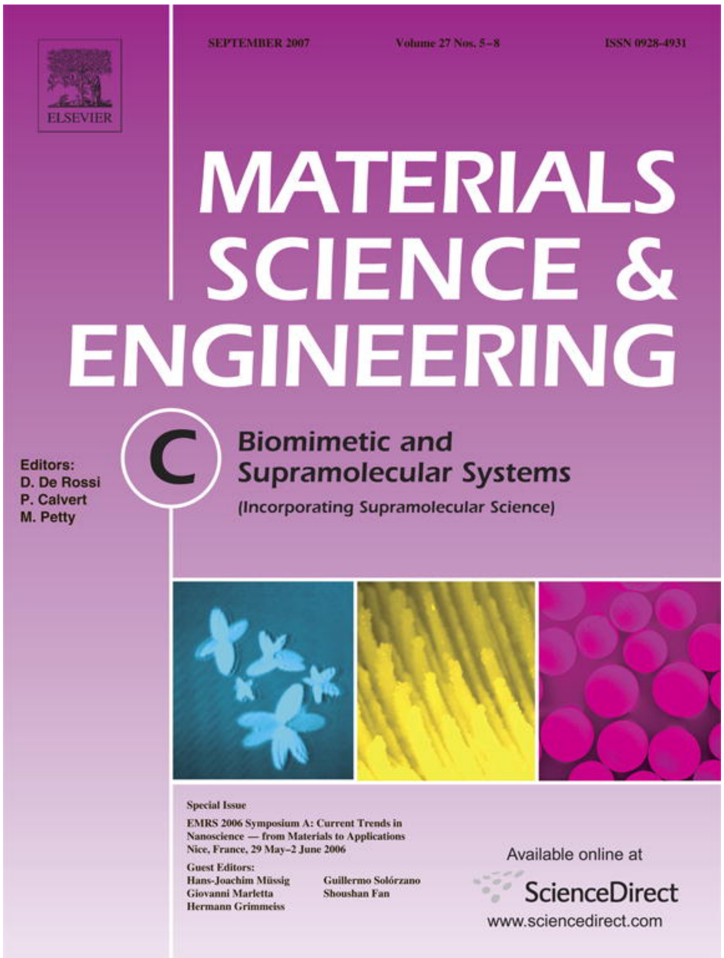
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Investigation of defect structure of InGaNAsSb/GaAs quantum wells

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Abstract

The results of the photoluminescence (PL) and the high-resolution X-ray diffraction (HRXRD) investigations of point and extended defects in strained InGaAs(N)Sb/GaAs quantum well (QW) structures grown at 478–505 °C are presented. HRXRD studies prove a good quality of heterointerfaces in all samples that is attributed to Sb-surfactant effect. The PL investigations show that the increase of the growth temperature of N-containing QWs leads to the increase of potential fluctuations in QW due to the increase of composition disorder. In the PL spectra an intense band caused by excitonic transitions related with N-related clusters in GaAs barriers is found. HRXRD mapping in symmetrical 004 reflections reveals the oscillation of interference picture in [110] direction around the normal to (100) surface known as a "wiggle". The mapping indicates the formation of elastically coupled domains which are elongated in [$\bar{1}$10] direction and are supposed to be cased by lateral composition modulations in the QW. It is proposed that a "wiggle" explained by the change of slopes of crystallographic planes with the depth is the result of competition of two factors — a symmetry of the surface stress tensor and a symmetry of bulk elastic moduli of a substrate material.

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1. Introduction

The rapid progress in optical fibre networks stipulates the barest necessity for the development of low-cost, un-cooled, thermally stable, vertical cavity surface emitting lasers (VCSELs) operating in the 1.3–1.5 μm spectral range [1]. Strained In(Ga)As/GaAs nanostructures are suitable candidates to succeed in the development of such devices. One of the possibilities is to apply coherently strained, self-organized three-dimensional islands (quantum dots) [2,3]. However, the problem of gain saturation in quantum dot structures is still not solved due to the difficulties in the production of quantum dots of identical sizes and shapes as well as of high density.

Another way is the use of highly strained InGaAs/GaAs quantum well (QW) structures [1]. But high lattice mismatch of InAs and GaAs results in the rapid degradation of optical quality of InGaAs QW when its transition wavelength is over 1.1 μm [4].

It has been found that adding of nitrogen in small amount (up to 3%) in the InGaAs decreases both lattice constant and band gap energy allowing to decrease a strain and to extend the emission wavelength of InGaAsN QW up to 1.3 μm [5–7]. However, it has been clearly demonstrated that the adding of only several percents of N into InGaAs can result in thickness and composition fluctuations in the QW and even in three-dimensional growth [8–11]. In order to obtain homogeneous alloys of InGaAsN, growth must occur under very low temperature metastable conditions that in its turn results in the increase of nonradiative defect density [12]. Nevertheless, recently low threshold InGaAsN/GaAs lasers beyond 1.5 μm have been obtained [13] by introduction of about 4% of N into $In_{0.4}Ga_{0.6}As$ QW at growth temperature of 350 °C.

One of the latest advances in the growth of InGaAsN alloys is the use of antimony as a surfactant [14–17]. It has been found that Sb-surfactant assisted growth of InGaAsN QWs leads to the

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increase of the photoluminescence intensity [14] and suppresses both plastic and elastic strain relaxation [15]. We have also shown [17] that the use of Sb as a surfactant expands the growth window of InGaAsN/GaAs QWs to higher temperatures without essential degradation of the photoluminescent characteristics. For further improvement of the optical properties of InGaAsNSb/GaAs QWs more information about the structural defects in this system is needed.

In this paper we present the results of the study of point and extended defects in GaAs-based heterostructures with strained InGaAsNSb and InGaAsSb single QW grown at relatively high temperatures (487–505 °C) by the low temperature PL and the HRXRD methods.

2. Growth procedure and experimental details

The structures studied were grown by molecular beam epitaxy (MBE) on (001) n-GaAs substrates using a V80H VG system equipped with In, Ga and Al effusion cells, As and Sb valved cracking cells, and an Addon radio-frequency (RF) plasma cell for N. Growth mode was controlled by *in situ* reflection high-energy electron diffraction (RHEED). The active region of the samples contained a 6 nm thick $In_{0.36}Ga_{0.64}As_{0.974}N_{0.016}Sb_{0.01}$ or $In_{0.36}Ga_{0.64}As_{0.99}Sb_{0.01}$ single QW grown on a 100 nm thick GaAs layer and followed by a 40 nm thick GaAs layer on the top. GaAs barriers were followed by 50 nm thick $Al_{0.4}Ga_{0.6}As$ layers. Each structure contained also GaAs buffer layer and a 50 nm GaAs cap layer. A schematic structure of the samples studied is presented in Fig. 1. AlGaAs and GaAs layers were grown at 590 °C and at As/III equivalent beam pressure ratio of 1.25. The InGaAsNSb QWs were grown at temperatures $T_{GR}=478$, 495, and 505 °C, while the reference InGaAsSb QW was grown at $T_{GR}=482$ °C. The N-containing samples were annealed under As vapour excessive pressure of $2\cdot10^{-6}$ Torr for 30 min at 730 °C in the MBE chamber immediately after the growth.

The N content in the InGaAsNSb QWs was estimated from the shift of 130 meV of room-temperature PL QW peak position in the sample grown at $T=478$ °C with respect to InGaAsSb one. In our previous study [17] this content was supposed to be ~1% in accordance with the shift of 150 meV/1% of N assumed for the structure with strain [5]. However, recent modelling within the effective-mass approximation and the two-level repulsion model have shown [18] that the modification of the band gap energies in the InGaAs epilayers and quantized energies in the InGaAs QWs due to N incorporation depends strongly on In content in the QW. Based on these calculations, the shift of 130 meV corresponds to introduction of ~1.6% of N in the $In_{0.36}Ga_{0.64}As$ QW.

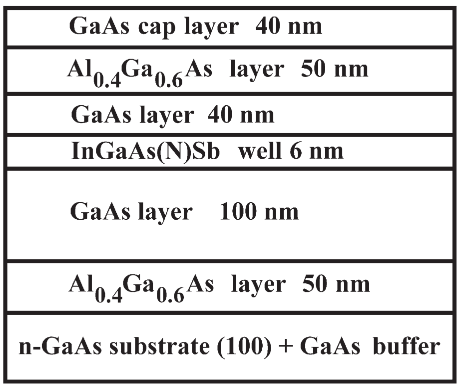

Fig. 1. Schematic structure of the samples studied.

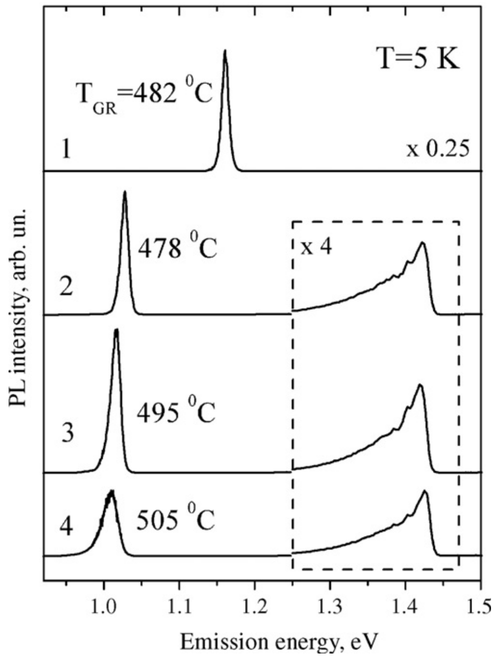

Fig. 2. PL spectra of the structures with $In_{0.36}Ga_{0.64}As_{0.99}Sb_{0.01}$/GaAs (curve 1) and $In_{0.36}Ga_{0.64}As_{0.974}N_{0.016}Sb_{0.01}$/GaAs (curves 2–4) single QWs grown at different temperatures: 482, 478, 495, and 505 °C (curves 1, 2, 3, and 4, respectively). $T=5$ K, $E_{exc}=2.41$ eV.

The PL spectroscopy was performed at temperature $T=5$ K. The PL was excited with 514.5 nm line of an Ar^+ laser with a ~5 W/cm^2 power density and detected by a cooled Ge detector with a standard lock-in technique. The HRXRD measurements were carried out using a high resolution X-ray diffractometer Philips MRD with a 4 × Ge (220) monochromator and Cu anode.

3. Results

Fig. 2 shows the low temperature (5 K) PL spectra of the samples studied. From the comparison of the PL spectra in the structures with InGaAsSb QW (curve 1) with respect to the N-containing ones (curves 2–4), it follows that adding of N results in the pronounced changes in the whole PL spectrum. Firstly, the QW peak position is red-shifted (Fig. 2, curves 1, 2) and continues to shift to the low-energy region when the QW growth temperature increases (Fig. 2, curves 2–4). Secondly, the integrated QW PL intensity decreases in 2.5–3.5 times in spite of post-growth annealing intended to decrease the density of nonradiative defects arising during N incorporation [12,19,20]. These defects are usually ascribed to N interstitials [19], as well as to ion-damage defects such as dislocations and clusters [20], As_{Ga}-related complexes [12] etc. At the same time the full width at a half maximum (FWHM) of the QW band increases

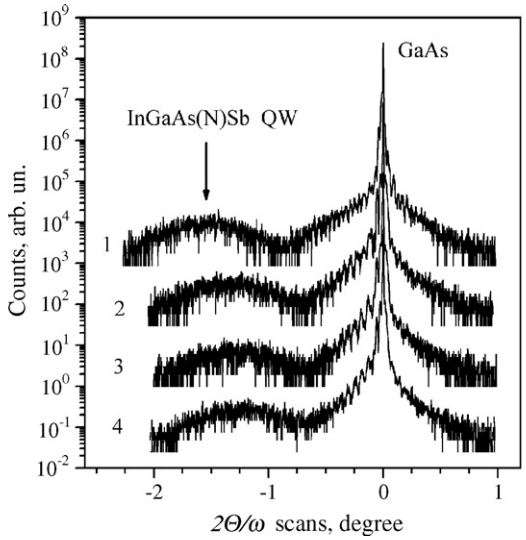

Fig. 3. HRXRD (004) rocking curves from the structures with $In_{0.36}Ga_{0.64}As_{0.99}Sb_{0.01}$/GaAs (curve 1) and $In_{0.36}Ga_{0.64}As_{0.974}N_{0.016}Sb_{0.01}$/GaAs (curves 2–4) single QWs grown at different temperatures: 482, 478, 495, and 505 °C (curves 1, 2, 3, and 4, respectively).

insignificantly from 10.5 meV (Fig. 2, curve 1) to 11.7 meV (Fig. 2, curve 2) when N is introduced at T_{GR} = 478 °C and continues to rise up to 24 meV (Fig. 2, curve 4) when T_{GR} increases to 505 °C.

Besides, in N-containing samples new PL band with maximum near 1.42 eV appears (Fig. 2, curves 2–4). It has an asymmetric shape with quite sharp high-energy edge and a stretched low-energy tail. We also observed this band in other structures with InGaAsNSb QWs with different In and N content and found that its intensity and spectral position do not depend on In or N content in the QW. It should be mentioned, that for our structures no growth interruption was done for the RF plasma ignition and running to maximum power. Since these processes need some time, the RF plasma was ignited when approximately half of GaAs barrier lying under the QW was grown. In spite of the shutter being closed at that moment, some N species can bypass through the shutter and incorporate in the GaAs barrier. In this case, the band near 1.42 eV can be caused by excitonic transitions related to the deep defects tentatively attributed to N-related clusters in GaAs [21,22]. Based on the compositional dependence of the low temperature PL spectra measured from the GaNAs alloys with low N content [22], we can estimate N content in GaAs barrier to be ~0.3%. Indeed, the shape and peak position of the 1.42 eV band is very similar to those observed in $GaN_{0.003}As_{0.997}$ [22].

HRXRD 2θ/ω (004) scans of the InGaAsSb QW and of the InGaAsNSb QW structures are presented in Fig. 3. In the curves the intense peak caused by GaAs substrate and GaAs layers as well as a broad peak of much lower intensity due to QW are observed. In the N-containing samples the QW-related peak is slightly shifted towards the GaAs peak indicating the decrease of lattice constant in the QW alloy. For all structures studied comparable interface quality is found. This is proved by sharp and well defined interference fringes observed in all scans.

To obtain more information about the strain and the defects in the structures the two-dimensional HRXRD maps of intensity distribution around the point of reciprocal lattice for symmetrical 004 and asymmetrical 113 and 224 reflections were studied. Fig. 4 presents the reciprocal space map close to the 004 reflection for the structure with InGaAsNSb QW grown at 495 °C. This map shows a surprising oscillatory intensity distribution along the normal to the crystal surface known as a "wiggle". These oscillations are observed in the region of thickness oscillations as well as in the region of the QW and GaAs. The effect of a "wiggle" has been found earlier [23] in the $In_{0.15}Ga_{0.85}As$ single QW

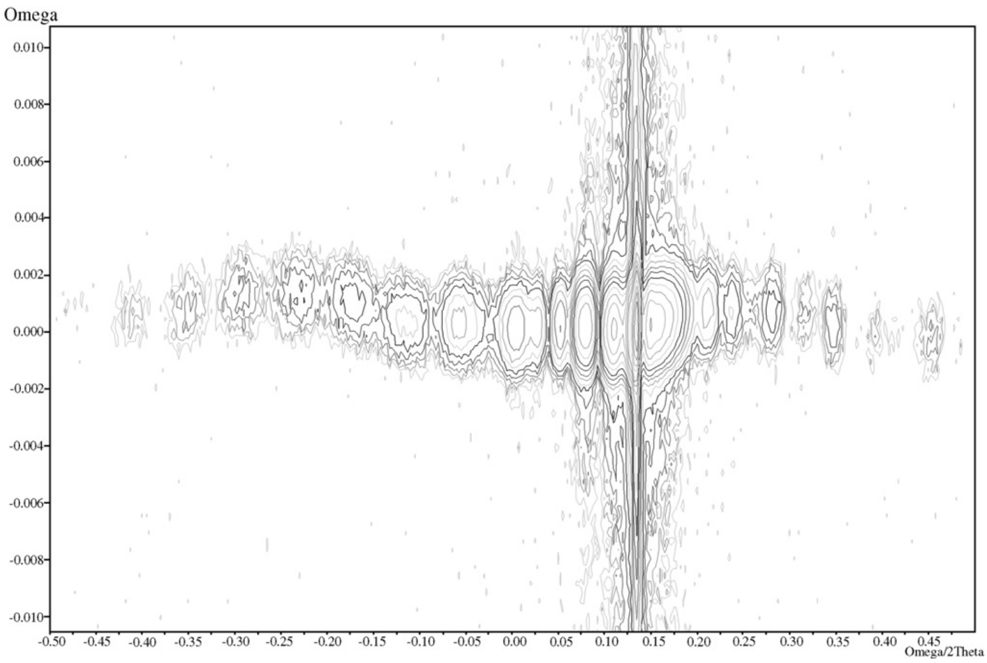

Fig. 4. Two-dimensional HRXRD maps for symmetrical 004 reflections of the structure with $In_{0.36}Ga_{0.64}As_{0.974}N_{0.016}Sb_{0.01}$/GaAs single QW grown at 495 °C.

embedded in the AlGaAs/GaAs structure. It has been mentioned there, that explanation of a "wiggle" is unknown, but it is supposed to be due to variation of slope of crystallographic planes as a function of depth.

We did not observe a "wiggle" in the reciprocal space maps for the asymmetrical 113 and 224 reflections in any structure studied. This indicates that the regions containing the misoriented crystallographic planes are not the surface layers but are located in deeper layers of the structure.

The azimuth investigations of the intensity distribution for 004 point revealed a "wiggle" for different directions. This means that misorientations of crystallographic planes have no single strongly pronounced direction. Moreover, in the ω-scan the intensity distribution in the azimuth direction [110] was found to be rather broad. In some structures, the ω-scan measured for the GaAs reflection revealed several peaks indicating the presence of some misoriented regions. The broadening was also observed both in the QW region and in the region of thickness oscillations.

4. Discussion

As it was mentioned above, the low temperature PL investigations reveal gradual red shift of the InGaAsNSb QW PL band position up to 18 meV with the T_{GR} increase. However, at room temperature the InGaAsNSb QW peak position changed only by 3–7 meV from sample to sample. A set of investigations [22,24,25] has shown that at low temperatures excitons in N-containing QWs are strongly localized on potential fluctuation of the band edge caused by local compositional disorder or interface roughness, while at room temperature excitons are delocalized. Therefore, we can suppose that the red shift of low temperature QW peak position is caused mainly by the increase of potential fluctuations in the QW. This is proved by the increase of FWHM as well as by the transformation of the shape of the QW band from almost symmetric in the 478 °C sample to asymmetric one caused by low-energy tail formation in the 505 °C structure. It is generally accepted that in dilute nitride semiconductors the low temperature band tail formed by disorder potential can be described by the exponential function [8,22,26]

$$g(E) = \frac{N_0}{E_0} \exp\left(\frac{E}{E_0}\right)$$

with some characteristic energy scale E_0 that is ascribed to localization potential of the fluctuations. Fitting of the low-energy tails to an exponential decay reveals the localization potentials of 4.2 meV in the InGaAsSb QW and of 5.1, 7.4, and 12.6 meV in the InGaAsNSb QWs grown at 478 °C, 495 °C, and 505 °C, respectively. In general, these potential fluctuations can be caused by composition fluctuations or by interface roughness.

As it was mentioned above, HRXRD investigations show well-defined interference fringes in all structures that proves a good quality of interfaces. The HRXRD data confirm that even in the 505 °C InGaAsNSb QW there are no considerable changes in the interface roughness. Thus, the increase of potential fluctuations in the QW with N adding as well as with the rise of growth temperature are caused mainly by the increase of composition fluctuations.

This conclusion is in agreement with the results of transmission electron microscopy (TEM) investigations of InGaAsN(Sb) QWs [9,11,14,15] that have shown a significant increase of composition fluctuations in the QW under N incorporation. It has been supposed [11] that these fluctuations are caused mainly by In and Ga non uniform distribution. The increase of the growth temperature of $In_{0.25}Ga_{0.75}As_{0.98}N_{0.02}$ QWs from 400 °C resulted firstly in the increase of composition fluctuations without changes of interface roughness and then at temperatures higher than 450 °C the evident undulations of the upper QW interface occurred [9]. In our structures quite sharp interfaces are observed in the structure with 505 °C QW that is obviously due to Sb-surfactant effect.

The analysis of HRXRD maps gives additional information about the defects in structures investigated. The specific feature of these maps is the presence of a "wiggle" that can be ascribed to variation of tilts of crystallographic planes with the depth [23]. The facts that this "wiggle" is observed in the region of QW, GaAs layers and of thickness oscillations while it is not observed for the asymmetrical 133 and 224 reflections give us a possibility to suppose that the variations in slope of crystallographic planes with the depth occur in the QW and in the adjacent barrier regions. Broadening of diffraction pattern in the map measured when diffraction plane is oriented along [110] direction indicates the formation in the QW of elastically coupled domains, which are elongated in $[\bar{1}10]$ direction. These domains can be caused by co-existence of phases of different surface reconstruction or lateral composition modulations [3]. Since the PL investigations show the presence of composition fluctuations in all QWs studied we think that these domains are composition modulations. This is in agreement with some high-resolution TEM investigations of the InGaAsN QWs [9,11] reported about the pseudo-periodic contrast in the QW with a periodicity of about ~10–20 nm due to local strain related to composition fluctuations. The formation of arrays of laterally periodic wire-like islands oriented along the $[0\bar{1}1]$ direction has been found in the InAs/GaAs (100) [27] and InGaAs/GaAs (100) [28] structures.

It is known that a symmetry of equilibrium periodical structures in the InAs/GaAs (100) heterosystem is determined by the interplay of two factors — a symmetry of the intrinsic surface stress tensor $\boldsymbol{\tau}_{ij}$ and a symmetry of bulk elastic moduli of zinc blende semiconductors [3,27]. For the (100) surface of zinc blende semiconductors, the directions of the main axes of the $\boldsymbol{\tau}_{ij}$ tensor are $[0\bar{1}1]$ and [011], while the bulk anisotropy of elastic moduli of these semiconductors is determined by the directions of the lowest stiffness [010] and [001]. The interplay of these two factors was supposed to be responsible for the change of orientation of the elastic domains along the $[1\bar{1}0]$ direction under submonolayer deposition of InAs on GaAs (100) to the orientation along the [100] and [010] directions under deposition of 1.0–1.5 monolayers of InAs [3]. We think that these factors are the reasons of appearance of a "wiggle". In fact, the increase of contribution of an energy of elastic strain caused by lattice mismatch of QW and substrate material in comparison with the contribution of an energy of surface strain on the heterointerface can result in the gradual change in slope of crystallographic planes from $[1\bar{1}0]$ to [100] directions with the depth of QW and perhaps of neighboring barrier layer that is a cause of a "wiggle".

5. Conclusion

Study of the structures with coherently strained InGaAs(N)Sb/GaAs single QWs by the PL and HRXRD methods proves that introduction of Sb results in good quality of heterointerfaces even in N-containing QWs grown at relatively high temperatures. However, it cannot prevent the formation of extended defects which manifest itself in HRXRD maps as the oscillation of interference picture in [110] direction around the normal to (100) surface known as a “wiggle”. A “wiggle” is observed in all structures studied and is supposed to be caused by the interplay of two factors — symmetry of the intrinsic surface stress tensor and symmetry of bulk elastic moduli of a substrate material that results in the change of slopes of crystallographic planes with the depth both in QW and in neighboring barrier layer. The latter is assumed to be connected with the presence of composition modulations in the QW proved by the PL investigations. Since a “wiggle” changes significantly the intensity of individual interference peaks in $2\theta/\omega$ (004) scans it must be taken into account during the simulation of measured scans.

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