In situ study of elastic strain relaxation in metamorphic InAs(Sb)/In(Ga,Al)As/GaAs heterostructures by using reflection high energy electron diffraction

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Abstract. We report on a comparative study of the elastic strain relaxation in InAs(Sb)/In(Ga,Al)As heterostructures grown by molecular beam epitaxy on GaAs substrates via InAlAs metamorphic buffer layer (MBL) with and without a highly strained 5 nm-thick GaAs insertion. Reflection high energy electron diffraction was used for in situ monitoring an in-plane lattice parameter of the epitaxial layers during growth. As a result, critical thickness of the initial stage of the InAlAs metamorphic buffer layer as well as its corresponding composition were determined for the structures with different position of the GaAs insertion within the MBL. The structure with 5 nm-thick GaAs layer inserted in the InAlAs MBL directly after achieving the In content of 37 mol.% demonstrates the highest room temperature photoluminescence at a wavelength of about 3.5 μm.

1. Introduction

High-efficiency emitters based on GaAs platform and operating in the mid-IR spectral region (2-5 μm) are of great demand due to variety of their applications including pollutant gas sensing, noninvasive disease analysis and IR-spectroscopy [1]. One of the main approaches to achieve this goal employs metamorphic technology, which allows fabrication of heterostructures with strongly mismatched In(As,Sb) active region on GaAs substrates via a metamorphic buffer layer (MBL) [2] providing a strain-relaxed layer of desired lateral lattice constant, namely, virtual substrate (VS). The residual strain accumulated in the MBL has a significant impact on structural and optical properties of the structures and is generally compensated by a certain inverse step representing the difference in composition between the MBL and VS [3]. However, besides the value of an inverse step, the most favorable MBL design is still unclear, and its MBE growth is attributed to a category of art [3]. Therefore, an in situ analysis of strain relaxation in the MBL is required to determine its optimized design, growth conditions and obtain low-defect-density metamorphic heterostructures with acceptable surface and structural quality. Such an analysis can be made by using the common in situ technique - reflection high energy electron diffraction (RHEED) - which allows one to measure directly the in-plane lattice parameter of a growing layer [4, 5].

In this paper, we report on in situ strain relaxation study of the convex-graded MBL InₓAl₁₋ₓAs (x = 0.05-0.87) of different design by using the RHEED technique.
2. Experimental procedure
The metamorphic InAs(Sb)/In(Ga,Al)As/GaAs heterostructures were grown by MBE using a RIBER 32P setup on GaAs(001) substrates via a convex-graded InAlAs MBL. In, Al, Ga fluxes were produced by conventional solid source effusion cells, whereas the As$_4$ flux was supplied from a VAC-500 valved cracking cell. The samples under the study contained sequentially from a GaAs substrate: a 0.2 μm-thick GaAs buffer layer, a 1.3 μm-thick In$_x$Al$_{1-x}$As MBL of grading composition $x = 0.05-0.87$ with a square root profile [6] and a 20 nm-thick In$_{0.75}$Al$_{0.25}$As VS layer. Details of the MBE growth can be found elsewhere [6]. The series of the samples differed from each other by the MBL design. Samples $B$ and $C$ contain a 5 nm-thick GaAs layer inserted into the MBL right after the point with In content of 21 mol.% and 37 mol.%, respectively. The same MBL without any insertion was used in a reference sample $A$.

IR pyrometer (IRCON) with a working temperature range of 350-700 °C was used to monitor a substrate temperature ($T_S$). In order to improve the reproducibility of $T_S$ setting, it was calibrated by using well-known temperatures of the oxides desorption from GaAs ($T_S = 580$ °C) and the (2×4)As-to-c(4×4) transition of GaAs surface reconstructions during the substrate cooling ($T_S = 510$ °C) under the As$_4$ flux immediately before the initiation of the MBL growth. In situ RHEED monitoring was used to observe surface reconstructions as well as to measure the in-plane lattice parameter of a growing layer.

For realization of the in situ RHEED technique we used a high-resolution Hamamatsu CCD camera C11440-50B with low noise, high readout speed and high dynamic range. We have developed original software to measure the in-plane lattice parameter ($a_∥$) with a high sensitivity of $10^{-4}$, being among the highest ever reported [5]. In the experiment, the RHEED intensity profile was recorded in the horizontal direction normal to the RHEED stripes (figure 1). To increase sensitivity and accuracy of the technique the profile has been averaged over a RHEED frame along the vertical axis. Then the distance $D$ between the main reflexes, which is inversely proportional to $a_∥$, was derived as the interval between the intensity maxima of the main reflexes on the horizontal profile. The experimental temporary dependence of $a_∥$ during the MBL growth was initiated each growth run at a GaAs buffer having the well known value of the lattice parameter.

Photoluminescence (PL) spectra at room temperature were measured under diode laser excitation ($\lambda_{exc} = 809$ nm) with a pumping power of 200 mW. Emitted light was detected with a nitrogen-cooled InSb photodiode and a standard lock-in amplifier.

![Figure 1](image-url)  
Figure 1. The RHEED (2×) pattern of GaAs (001) surface (right) and the profile of the RHEED intensity measured in the horizontal direction (left). $D$ is the distance between the maxima of the main RHEED reflexes.
3. Results and discussion

The temporal dependence of the in-plane lattice parameter during growth of sample A is shown in figure 2. The plot demonstrates that the lattice parameter increases with the In content rising non-monotonously. Moreover, a sharp rise of $a_\parallel$ in the initial stage of the MBL growth (marked by a vertical arrow in figure 2) is clearly seen. It obviously evidences the onset of the strain relaxation process in the MBL. Taking into account the growth rate (0.7 ML/s) and the In content-vs-thickness square root profile $x(d)$, the critical thickness ($d_{cr}$) and the corresponding critical In composition ($x_{cr}$) can be easily obtained. One can see also multiple small steps on the curve that indicate a stepwise mechanism of strain relaxation in MBL [7].

![Figure 2. The temporary dependence of the in-plane lattice parameter measured during the growth of sample A. A vertical arrow marks the onset of strain relaxation in the MBL.](image)

| Sample | A         | B         | C         |
|--------|-----------|-----------|-----------|
| Critical thickness ($d_{cr}$), nm | 85 ± 5    | 85 ± 5    | 100 ± 5   |
| Critical In content ($x_{cr}$)  | 0.22 ± 0.02 | 0.22 ± 0.02 | 0.28 ± 0.02 |

Table 1. The critical thicknesses and compositions of the sample studied.

Figure 3 shows the initial parts of the dependencies of $a_\parallel$ vs layer thickness, measured for samples A, B and C. The critical thickness of the initial part of the convex-graded MBL employed has been found to be accurately reproduced for different growth runs. It is obviously the case for sample B.
which contains the 5 nm-thick GaAs layer inserted in the MBL at a thickness exceeding appreciably the critical one (see $d_{cr1}$ for curves 1 and 2 in figure 3). On the other hand, the incorporation of the thin GaAs layer with a smaller lattice parameter in the MBL region before the crucial point results in partial release of the accumulated strain and substantial enlargement of the critical thickness (see $d_{cr2}$ for curve 3 in figure 3). The parameters $d_{cr}$ and $x_{cr}$ derived for the samples studied are presented in table 1. It should be noted that the value $d_{cr1}$, despite it was obtained for the graded In$_x$Al$_{1-x}$As layer ($x = 0.05-0.22$), is comparable with that measured for a layer of constant composition In$_{0.05}$Al$_{0.95}$As [8]. We believe that it is not surprising if we remember that the $d_{cr1}$ value was measured at growth temperature and could be increased after the sample cooling down to room temperature due to additional relaxation of stress.

![Figure 3](image_url)

Figure 3. The dependencies of the in-plane lattice parameter on the thickness of the growing MBL for samples A (1), B (2) and C (3). $d_{cr1}$ and $d_{cr2}$ are the critical thicknesses obtained for samples A, B and C, respectively.

We have also investigated PL properties of metamorphic InAs(Sb)/InGaAs/InAlAs quantum well (QW) heterostructures grown atop MBLs of the same design as was used in the above studied reference samples. The InAs(Sb)/InGaAs/InAlAs QW active region was the same for all the structures studied and its design can be found elsewhere [9]. The PL spectra of the structures measured at room temperature are shown in figure 4. One can see that the PL intensity at a wavelength of $\sim$ 3.5 μm of the structure with GaAs interlayer inserted in the MBL at a thickness beyond $d_{cr1}$, exceeds about twice that of the structure with the reference MBL without GaAs insertion. In contrast to that, the structure with the GaAs interlayer inserted in the MBL at thickness below $d_{cr1}$ demonstrates no PL in the interested
such behavior correlates well with the residual strain level in the MBL. The higher the residual strain in the MBL, the higher is the probability of the strain relaxation in the QW active region grown atop of the MBL. The strain relaxation is well known to be accompanied by generation of misfit and threading dislocations which, in turn, result in the PL intensity drop. The highest and the lowest level of residual strain are observed in samples C and B, respectively. It can be easily seen from the dependences in figure 3, keeping in mind that accumulation of residual strain in the MBL results in decreasing $a_{||}$, and the highest and the lowest $a_{||}$ values at the same thickness (or In content) are observed in sample B and sample C, respectively.

![Figure 4: The room temperature PL spectra of the InAs(Sb)/InGaAs/InAlAs QW structures grown atop of MBLs of different design: 1 - as in sample A, 2 - B, and 3 - C.](image)

### 4. Conclusions

In the paper, the strain relaxation in the convex-graded In$_x$Al$_{1-x}$As ($x = 0.05-0.87$) MBLs with and without a 5 nm-thick GaAs insertion were studied by using in situ RHEED picture analysis. The critical thicknesses and the corresponding In contents of the initial part of the MBLs were derived. The incorporation of the GaAs insertion into the MBL right after the In content in InAlAs achieves 37 mol.% has been shown to result in doubling the room temperature PL intensity ($\lambda \sim 3.5 \, \mu m$) from the InAs(Sb)/InGaAs/InAlAs QW structure grown atop of the MBL.
Acknowledgments
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References
[1] Shterengas L, Kipshidze G, Hosoda T, Liang R, Feng T, Wang M, Stein A and Belenky G 2017 Cascade pumping of 1.9–3.3 μm type-i quantum well GaSb-based diode lasers IEEE Journal of Selected Topics in Quantum Electronics 23 1-8
[2] Choi H, Jeong Y, Cho J and Jeon M H 2009 Effectiveness of non-linear graded buffers for In(Ga, Al) As metamorphic layers grown on GaAs(001) J. Cryst. Growth 311 1091-5
[3] Chernov M Y, Solov’ev V A, Komkov O S, Firsov D D, Meltser B Y, Yagovkina M A, Baidakova M V, Kop’ev P S and Ivanov S V 2017 Enhanced room-temperature 3.5 μm photoluminescence in stress-balanced metamorphic In(Sb,As)/In(Ga,Al)As/GaAs quantum wells Applied Physics Express 10 121201
[4] Munekata H, Chang L L, Woronick S C and Kao Y H 1987 Lattice relaxation of InAs heteroepitaxy on GaAs J. Cryst. Growth 81 237-42
[5] Koblmüller G, Averbeck R, Riechert H, Hyun Y J and Pongratz P 2008 Strain relaxation dependent island nucleation rates during the Stranski–Krastanow growth of GaN on AlN by molecular beam epitaxy Appl. Phys. Lett. 93 243105
[6] Chernov M Y et al 2017 InSb/InAs/InGa(Al)As/GaAs(001) metamorphic nanoheterostructures grown by MBE and emitting beyond 3 μm J. Cryst. Growth 477 97-9
[7] Song Y, Wang S, Tångring I, Lai Z and Sadeghi M 2009 Effects of doping and grading slope on surface and structure of metamorphic InGaAs buffers on GaAs substrates Journal of Applied Physics 106 123531
[8] Shan R, Liu Y, Guo J, Wang G and Xu Y 2016 Growth and characterization of high strain InGaAs/GaAs quantum well by molecular beam epitaxy Infrared Technology and Applications, and Robot Sensing and Advanced Control 10157 101573F
[9] Solov’ev V A, Chernov M Y, Komkov O S, Firsov D D, Sitnikova A A and Ivanov S V 2019 The Effect of Highly Constrained GaAs and InAs Inserts in the InAlAs Buffer Layer on the Structural and Optical Properties of Metamorphic InAs(Sb)/InGaAs/InAlAs/GaAs Quantum-Size Heterostructures JETP Letters 109 377-81