

Heterointerface width in InGaAs/InAlAs/InP heterostructures for quantum cascade lasers grown by molecular beam epitaxy

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The width of heterointerfaces in strain-compensated $\text{In}_{0.36}\text{Al}_{0.64}\text{As}/\text{In}_{0.67}\text{Ga}_{0.33}\text{As}$ heterostructures for quantum cascade lasers grown by molecular beam epitaxy on InP (001) substrates with various-thickness $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layers was investigated by X-ray reflectometry. It was found that reducing the buffer layer thickness from 500 to 100 nm does not degrade the quality of the formed heterostructures and maintains the heterointerface width at a level not exceeding one monolayer (≤ 0.35 nm).

Keywords: quantum cascade laser, heterostructure, heterointerface width, molecular beam epitaxy, X-ray reflectometry.

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Roughness of heterointerfaces in InGaAs/InAlAs/InP heterostructures of mid-IR quantum cascade lasers (QCLs) on InP substrates [1,2] can have a significant impact on the QCL quantum efficiency and threshold current [3,4]. This is because of enhancement of charge carrier scattering at heterointerfaces and violation of the coherence of radiative electron transitions in QCL heterostructures. In addition to roughness, heterointerfaces may have a heterojunction gradient caused by mixing of the interface materials due to their mutual diffusion and/or in the process of growing. Such a heterojunction gradient promotes formation of a blurred the profile of potential barriers and wells in the energy band diagram thus inducing variations in the energies of size quantization levels [5] different from those calculated for a sharp-heterointerface configuration. As a result, this may affect the differential efficiency and threshold current [6], and also cause deviation of the QCL emission wavelength from the designed one [7]. Roughness and heterojunction gradient jointly determine the heterointerface widths and modify QCL functional characteristics; therefore, atomic-level control of the heterointerface and minimization of its width is critical for improving the QCL performance.

An important step in the epitaxial process is the substrate pre-treatment, that is, removing natural oxide from the substrate surface via thermal annealing. However, annealing increases the substrate roughness [8]; therefore, prior to epitaxial growth of the heterostructure functional layers, it is necessary to create a buffer layer ensuring surface flattening. In our previous study [9] we considered the influence of the InP substrate morphology on the roughness of heterointerfaces of QCL heterostructures grown on

InP buffer layer pre-grown on the InP (001) substrate by metal-organic vapor phase epitaxy (MOVPE). It was shown that, during MBE-growth of QCL heterostructures on a 50 nm thick $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layer, roughness of the quantum cascade heterointerfaces depends on morphology of the InP substrate surface. Paper [10] also shows that, when the GaAs layer is grown on the GaAs substrate by MOVPE, there is observed a significant dependence of the epitaxial layer roughness on the initial substrate roughness and surface precleaning from oxides. On the other hand, paper [11] shows that, in growing a GaAs buffer layer on GaAs substrates by MBE method, a considerable reduction of the root-mean-square surface roughness is observed; this reduction takes place at the GaAs layer thickness of 100 nm, while at the thickness of 500 nm the surface roughness is fully independent of the substrate morphology.

Since the heterointerface width is determined by accounting for both the heterojunction gradient controlled by selecting the epitaxial growth conditions and heterointerface roughness affected by the morphology of the epitaxial growth starting surface, it is necessary to ensure, under the selected epitaxial growth parameters, planarization of the buffer layer surface that depends on the layer thickness. The goal of this work was studying the effect of thickness of the $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layer grown on the surface of InP (001) substrate 50.8 mm dia upon the thickness of heterointerfaces of strain-compensated QCL $\text{In}_{0.36}\text{Al}_{0.64}\text{As}/\text{In}_{0.67}\text{Ga}_{0.33}\text{As}$ heterostructures 0.5 μm in total thickness grown immediately upon the buffer layer formation. In such heterostructures, the selected thickness of the $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layer whose refraction index

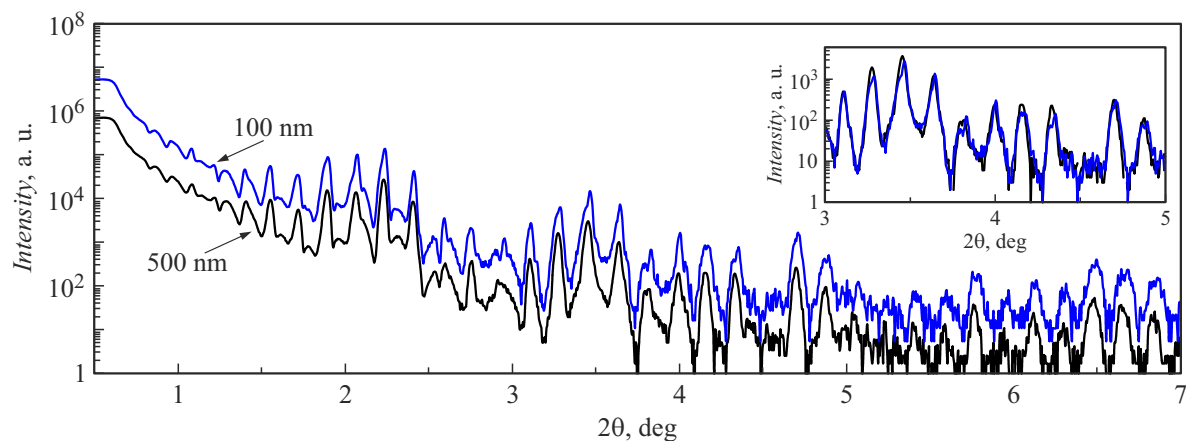


Figure 1. X-ray reflectometry curves for samples *B100* and *B500*. For clarity, the curves are vertically shifted relative to each other. The inset presents a scaled-up fragment with the curves aligned in intensity.

differs from that of the InP substrate affects the lightwave distribution in the QCL waveguide and, hence, affects the QCL characteristics [12]. In this connection, determination of the minimum sufficient buffer layer thickness ensuring the surface flattening and meeting the requirements for lightwave distribution in the QCL waveguide is of great practical interest. This study aims to solve this problem by growing and analyzing heterostructures with buffer layers of different thicknesses.

Heterostructures examined in this study were grown by MBE using InP (001) substrates with epi-ready polished surfaces. To remove natural oxide from the substrate surface prior to the epitaxial growth, pre-annealing at $300 \pm 10^\circ\text{C}$ was performed and followed by high-temperature annealing at $530 \pm 10^\circ\text{C}$ for 10 min. During this time, the natural oxide gets removed from the surface of the semiconductor wafer, and the substrate surface is modified by forced grinding and polishing. This modification manifests itself in formation of a relief on the wafer surface, which is accompanied by appearance of a pattern of point high-energy electron diffraction. The pattern shows that the surface is not smooth, i.e. roughness is present.

The epitaxial process started with formation of the $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layer 100 or 500 nm thick (samples *B100* or *B500*, respectively). During the buffer layer growth, the surface got flattened, which was confirmed by emergence of characteristic bands in the high-energy electron diffraction pattern. After that, there was formed the QCL active region 500 nm in total thickness consisting of ten cascades of alternating layers of $\text{In}_{0.36}\text{Al}_{0.64}\text{As}/\text{In}_{0.67}\text{Ga}_{0.33}\text{As}$ solid solutions with layer thicknesses of 1.3–4.3 nm and total cascade thickness of 50.4 nm. The active region design is described in more detail in [9]. On top, a capping $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ layer 50 nm thick was grown. The heterostructures' heterointerface widths were examined by small-angle X-ray reflectometry (XRR) using diffractometer PANalytical X'Pert PRO MRD in the high-resolution configuration with a primary-beam hybrid

monochromator consisting of a parabolic X-ray mirror and a Ge (400) 4-bounce monochromator ($\text{CuK}_{\alpha 1}$ radiation). The curves were measured by 2θ – ω -scanning with narrow slits on incident and reflected beams of specular-reflection from the pre-aligned sample.

X-ray reflectometry is based on analyzing the dependence of specularly reflected X-ray radiation intensity on incidence angle ω . The reflectometry curve provides information on the depth profile of electron density averaged over the sample plane, which enables determination of the material thickness and density, as well as of layer heterointerface widths. This is why the X-ray reflectometry (XRR) method, in contrast to, e.g., the widely used method of atomic force microscopy [13], determines the total heterointerface width that includes both the interface roughness and mixing of materials. Since both of these factors negatively affect the QCL properties, XRR may be regarded as a more informative research method.

The results of XRR measurements versus radiation reflection angle 2θ for the *B100* and *B500* samples are shown in Fig. 1. Coincidence of the peaks' angular positions evidences that the quantum cascade layer thicknesses do not differ, which confirms good reproducibility of sequentially grown heterostructures. In the inset to Fig. 1, XRR curves for samples *B100* and *B500* are aligned in intensity. An increase in the heterointerface width significantly reduces peak intensities in the XRR curve, which becomes most pronounced when the scanning angle increases [9]. The almost full coincidence indirectly indicates close values of the heterointerface widths in two heterostructures under consideration.

To quantify heterointerface widths, the XRR curves were simulated via the Parratt formalism [14] by using the Nevo–Kroce heterointerface model [15]. In the course of numerical simulation, the best agreement between the calculated and experimentally measured curves was achieved by varying thicknesses of the $\text{In}_{0.36}\text{Al}_{0.64}\text{As}/\text{In}_{0.67}\text{Ga}_{0.33}\text{As}$ layers and root-mean-square heterointerface width. The

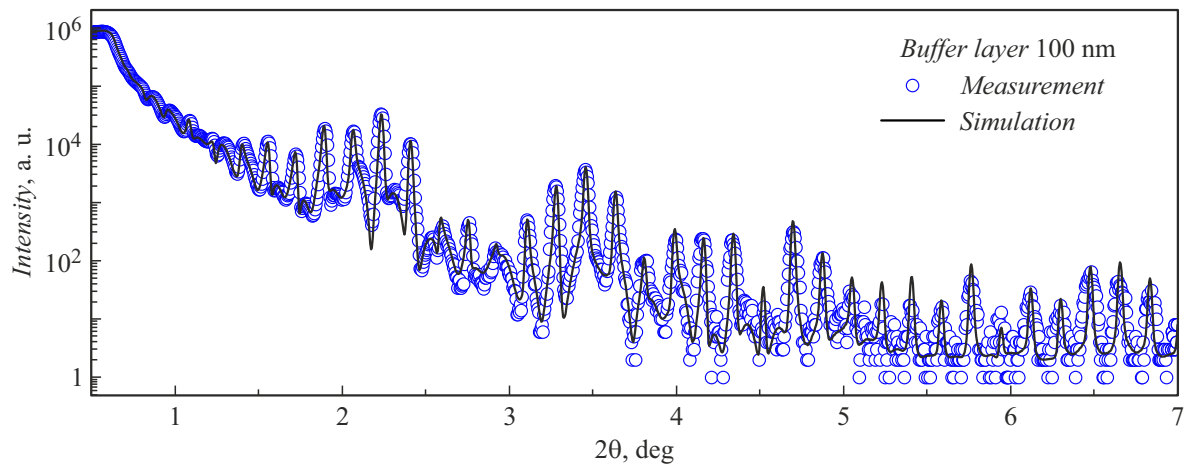


Figure 2. Simulated and measured XRR curves for the B100 sample.

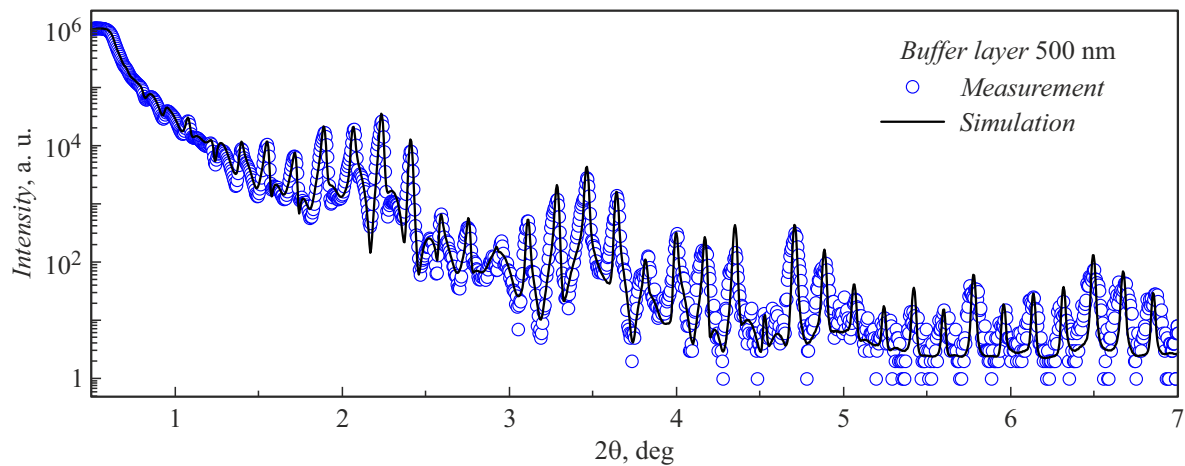


Figure 3. Simulated and measured XRR curves for the B500 sample.

Simulation of the X-ray reflectometry curves

Epitaxial layer	Heterointerface width, nm	
	Sample B100	Sample B500
InAlAs	0.33 ± 0.09	0.35 ± 0.08
InGaAs	0.29 ± 0.07	0.30 ± 0.07

heterointerface widths were set equal for all the periods. Figs. 2 and 3 illustrate the comparison of measurement and simulation results, which demonstrates their good agreement at the heterointerface widths given in the Table.

The simulations show (see the Table) that widths of the quantum cascade heterointerfaces in the considered heterostructures is 0.33–0.35 and 0.29–0.30 nm for the InAlAs/InGaAs and InGaAs/InAlAs heterointerfaces, respectively. Thicknesses of the heterostructure cascades with the thinnest layer thickness of 1.2 nm were 49.3 and 49.2 nm for the B100 and B500 samples, respectively.

The obtained results show that differences in heterointerface widths and cascade thicknesses of different samples are insignificant (small as compared to the material lattice constant); this indicates high reproducibility of the MBE technique and stability of the QCL heterostructures growth. Important epitaxial process parameters that allow achieving the mentioned effect are the epitaxy temperature and flow of the group V elements. In our case, when InGaAs and InAlAs layers with high indium content are grown, the optimal epitaxial temperature ($490 \pm 10^\circ\text{C}$) is below the substrate pre-annealing temperature. Therewith, to increase the mobility of group III atoms on the epitaxial surface, a relatively low ratio of flows of group III and group V elements (1 to 3) is used. As a result, it is possible to create multilayer heterostructures with heterointerface widths of about one atomic monolayer. To ensure uniformity of the epitaxial layer thickness over the wafer area and identity of the layer thicknesses in each QCL heterostructure cascade, MBE setups apply rotation of the semiconductor wafer carrier. In growing thick layers when minor deviations

in thickness are acceptable, the rotation speed is selected to be low: high rpm accelerates the wear of expensive bearings of the high-vacuum reactor. However, the rotation speed should be increased in growing QCL heterostructures with the quantum cascade layer thickness of 1–2 nm and growth duration of 5–10 s. In our case, rotation speed was increased to 40 rpm (the typical value is 20 rpm) in order to ensure higher uniformity.

Keep in mind that the depth of X-ray penetration into the sample in the process of XRR examination depends on incidence angle ω , i.e. it increases with increasing angle 2θ on the XRR curve. In the range of scanning angles used in this study, radiation penetrates into the heterostructure to a depth of about 1 μm and reaches the substrate. Measuring by XRR implies averaging the heterointerface widths over the irradiated area but not over the depth, since the scattered radiation is summed up taking into account the phase (but not simply over intensity, as when summing over the area). In simulating the XRR curves we assumed that the heterointerface width did not decrease or increase during the QCL heterostructure growth, i.e., it remained constant from the buffer layer to the surface. This is a quite realistic scenario in the case of a proper growth mode. The variable heterointerface width can manifest itself on the XRR curve either as a strong deterioration of contrast and decrease in oscillation amplitude with increasing angle 2θ or, conversely, in low informativeness of the XRR curve at small angles and increase in the contrast of oscillations with increasing 2θ . This effect is not observed in the experimental XRR curves, which allows us to confidently speak about the heterointerface width constancy throughout the growth of entire heterostructure. In our case, this is also confirmed by a good agreement between the simulated and experimental curves.

The absence of a significant dependence of the widths of active-region heterointerfaces on the buffer layer thickness ranging from 100 to 500 nm allows making an important practical conclusion that, already at the buffer layer thickness of 100 nm, the substrate morphology that has degraded during thermal pre-annealing gets completely levelled. A slight excess of the InAlAs heterointerface width over the InGaAs width (see the Table) is within the accuracy and may be caused by differences in kinetics of surface processes and adatom migration rates for these materials under the MPE conditions.

Paper [9] shows that formation of a thick InP buffer by MOVPE followed by MBE growth of a 50 nm thick InGaAs layer does not provide the required levelling of the substrate morphology effect. The results obtained in this work for the MBE-grown InGaAs buffer layers 100 and 500 nm thick are generally consistent with those of [11] that have demonstrated the efficiency of using MBE for creating a buffer layer in the GaAs/GaAs system. The determined heterointerface widths in the heterostructures under study are several times smaller than that of the thinnest active-region layers (1.2 nm); due to this, the

QCL active region may be fabricated with high precision matching the designed configuration of energy levels. Note that, in the case of MOVPE-grown heterostructures, heterointerface widths determined by secondary ion mass spectrometry may be 2.5–4.5 nm [7]. A similar method was used in [16] for MOVPE-heterostructures to demonstrate a smaller heterointerface width which, as per the Ga and Al concentration profile, amounted to about 0.5 nm; however, the In concentration profile was more blurred and gave the heterointerface width of about 1 nm. The blurred profile of the In content revealed by secondary ion mass spectrometry is determined, e.g., by the In atoms segregation manifesting itself for both the MOVPE and MBE growths. However, work [6] has shown that segregation is more pronounced in growing by MOVPE and is, for instance, a possible reason for broadening of the cathodoluminescence emission spectrum with respect to the case of growing heterostructures by MBE. By considering the influence of heterointerface parameters on the laser output characteristics, the authors of [17] revealed relatively large values of roughness, as well as of heterojunction gradient width in MOVPE-grown QCL heterostructures (0.55 nm) compared to those in the case of MBE growth (0.4 nm). Significant difference in record values of room-temperature efficiency of MPE-grown QCLs (31 %) [18] and MOVPE-grown QCLs (19.1 %) [17] is also indicative. Based on the cited results, as well as on the results of this study, we may state that MBE is more preferable technique for creating QCL active-region heterostructures.

Thus, using X-ray reflectometry and simulation of reflectometry curves, we have analyzed the heterointerface widths of strain-compensated $\text{In}_{0.36}\text{Al}_{0.64}\text{As}/\text{In}_{0.67}\text{Ga}_{0.33}\text{As}$ heterostructures of quantum cascade lasers formed by MBE on various-thickness $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layers grown on InP (001) substrates. The studies have shown that the buffer layer thickness of both 100 nm and 500 nm ensures, under the specified epitaxial conditions, subsequent formation of quantum cascade laser heterostructures with heterointerface widths of no more than 0.35 nm. The obtained heterointerface widths correspond to the one-monolayer material thickness and clearly demonstrate the advantages of growing the QCL active region by MBE over MOVPE.

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Conflict of interests

The authors declare that they have no conflict of interests.

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