Molecular-beam epitaxy growth and characterization of 5-μm quantum cascade laser

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Abstract. Molecular-beam epitaxy growth of 5 μm emitting strain-compensated quantum semiconductor laser (QCL) is reported. The QCL structure is characterized by complementary techniques: high-resolution X-ray diffraction and dynamical secondary-ion mass-spectrometry, that reveal the high quality of QCL structure and in-depth distribution of chemical composition, respectively.

1. Introduction
The pioneering work on quantum cascade lasers was first done by Kazarinov and Suris in 1971 [1] describing what would later be called a ‘diagonal’ or ‘photon-assisted’ tunneling. Faist and Capasso demonstrated the first intersubband laser, designed to emit at 4.3 μm wavelength and grown by molecular beam epitaxy (MBE) in 1994 [2]. They named new laser as quantum cascade laser (QCL). Progress advanced quickly after that and is mostly due to the understanding of how bandstructure engineering, pioneered by Capasso, can be most successfully used to control the electron flow and thus increase population inversion and modal net gain, which ultimately controls the laser threshold [3]. After the first attempts the so-called ‘three-well vertical’ design was established in a wavelength range from 5 to 11 μm [4] and more, and the first far-infrared QCL with wavelength larger than 80 μm (87 and 130 μm) were demonstrated [5]. The wavelength is a function of an application for different QCL and one of the application is using atmospheric windows at 3 - 5 μm and 8 – 13 μm for very high speed communication, with satellites also, without turbulence and absorption as in near infrared and visible spectrum ranges [3] (see Figure 1).

In this paper we report the formation and characterization of QCLs for 5 μm range lasing based on AlInAs/GaInAs lattice matched superlattices grown by MBE on the InP substrate with ‘three-well’ design of its active zone. Such design allows to achieve both the shortest ground state life time (~0.4 ps) and high injection efficiency [6].
2. Experimental Procedure

The QCLs investigated here were grown by MBE in RIBER 32 system. The growth temperatures were 500°C, and slightly As-stabilized conditions were applied for all samples. The whole layer structure was designed following [6] and was deposited onto n-InP:Sn substrate which was doped only at a level of $2 \times 10^{17}$ cm$^{-3}$. Active regions of AllnAs/GalnAs heterostructures were grown in a lattice matched mode. The active region of the QCL structure consisted of 25 periods. One period (Figure 2) constitutes a sequence of 18 alternating AllnAs/GalnAs layers in growth direction and is composed as
follows (thicknesses in Å): 30/36/28/45/10/27/13/23/17/20/16/19/21/21/21/21, starting with the AlInAs exit barrier where the underlined numbers represent well layers and the bold numbers indicate n-doped layers, that were doped with Si at a level of $4 \times 10^{17}$ cm$^{-3}$.

The whole structure consists, from substrate to top, of the digitally graded AlGaInAs (Si, $2 \times 10^{17}$ cm$^{-3}$, 30 nm thickness) layer, the GaInAs (Si, $1 \times 10^{17}$ cm$^{-3}$, 300 nm thickness) waveguide, the active region (thickness 1135 nm) consisting of 25 periods, the GaInAs (Si, $1 \times 10^{17}$ cm$^{-3}$, 300 nm thickness) waveguide, the AlGaInAs (Si, $2 \times 10^{17}$ cm$^{-3}$, 30 nm thickness) digitally graded layer, the AlInAs (Si, $2 \times 10^{17}$, $3 \times 10^{17}$, and $7 \times 10^{18}$ cm$^{-3}$, 2500 nm thickness) cladding layer, the AlGaInAs (Si, $7 \times 10^{18}$ cm$^{-3}$, 30 nm thickness) digitally graded layer, and the GaInAs (Si, $2 \times 10^{19}$ cm$^{-3}$, 10 nm thickness) top contact layer. Each period comprises of a three quantum-well (QW) active region and a graded-gap superlattice injection region (Figure 2). The whole structure was grown following [7] for achieving 5 μm laser generation. Finally, QC lasers was fabricated emitting around 5 μm.

The QCL structure was characterized by high-resolution X-ray diffraction (HRXRD) and dynamical secondary-ion mass-spectrometry (D-SIMS) to reveal the crystalline quality and the chemical composition, respectively.

The HRXRD was studied using a D8 Discover X-ray diffractometer (Bruker AXS, Germany) in a double-crystal arrangement with a channel-cut analyzer. The source of X-ray radiation was a tube with a rotating copper anode of power of 6 kW. The HRXRD curves were obtained by $2\theta/\theta$ scanning in the region of the reflection angle from the InP (004).

D-SIMS was carried out using the CAMECA IMS-7f secondary ion microprobe. In-depth profiling of matrix elements was done using Cs$^+$ primaries with an impact energy of 3 keV and CsX$^+$ (X=Al, Ga, In, and As) secondary ions. High depth resolution profiling of the QCL active region was done using O$_2^+$ primaries with an impact energy of 2 keV impinging at an angle of 45°. The depth distribution of silicon was measured using Cs$^+$ primaries while following the Si$^-$ secondaries under high-mass resolution mode M/ΔM = 3000 to avoid mass interference of the $^{28}$Al$^1$H$^-$ and $^{28}$Si$^-$ analytical signals. The depth of the sputtering crater was measured by mechanical stiulus profilometer AMBIOS XP-1.

3. Results and discussion

Figure 3 shows the SEM image of the mesa surface of the QCL structure under post-treatment preparation. One can see distinctly 25 periods of the active zone with waveguides around it.
The multiple mode lasing spectra of the QCL above the threshold are shown on Figure 3 for 78 K and 300 K operating temperatures. The QCL emits at a wavelength of 5.05 \( \mu \)m and 5.24 \( \mu \)m at 78 K and at 300 K, respectively. The lasing characteristics of the QCL been described in detail elsewhere [7].

The diffraction curve for QCL sample is shown in Figure 4. On the curve an intense central peak is observed. The location of this peak corresponds to the Bragg diffraction from the InP substrate. In addition to this peak, the developed pattern caused by the structure of the active region S1 is observed in curve. The period in the spatial arrangement of active region S1, which was calculated from the distances between interference fringes, is equal to 470±20 Å. This result is in good agreement with the SIMS profiling data.

As seen from insert on Figure 4 the central peak of the interference pattern S10 as well as peaks corresponded AlInAs and GaInAs layers are overlapped with the peak of InP. It means that these layers show acceptably low lattice-mismatch \( \Delta c/c \). The \( \Delta c/c \) is equal to 1.2\( \times 10^{-3} \), 4.0\( \times 10^{-4} \), and -8.7\( \times 10^{-4} \) for AlInAs, GaInAs and S10 (or GaAlInAs), respectively.

![Figure 3. SEM image of the QCL mesa stripe under processing.](image_url)
From the values of Δc/c we can adjust composition of the upper layers as well as average one in the SI. The initial compound was taken from the technological data. The molar concentration of InAs in AlInAs and GaInAs layers are equal to 0.528 and 0.524, respectively. The molar concentration of GaAs and AlAs, in the SI GaAlInAs layer forming the active region is equal to 0.21 and 0.27, respectively. It should be noted that the concentration obtained from the analysis of X-ray diffraction is in good agree with the values obtained from SIMS profiling.

A well developed interference patterns indicates a high planarity of the interfaces in the active region of the QCL structure. The inset (2θ ≈ 63.3°) shows the curve recorded in high-resolution mode to separate substrate and SI0 peaks.

Al, Ga, In, As and Si depth profiles are shown in Figure 5. The results of high depth resolution SIMS profiling of the active region are presented in Figure 6. The quantification SIMS procedure for chemical composition was done using the set of single-layer epitaxial structures as external standards. Quantification SIMS procedure for in-depth distribution of the silicon dopant was done using implanted standards according to the well defined SIMS protocol [8].

The in-depth distributions of the chemical composition and distribution of donor atoms revealed by D-SIMS profiling are in good agreement with the structure of QCL.
Figure 5. SIMS profiles of epitaxial QCL device structure of matrix elements and the donor impurity.

Figure 6. High-depth resolution SIMS profiles of matrix elements in the active region of QCL

4. Conclusions
Molecular-beam epitaxy was used to fabricate strain-compensated quantum cascade lasers emitting at a wavelength of 5 μm. The quality of the QCL structure was characterized by high-resolution X-ray diffraction and dynamical SIMS in-depth profiling.

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