MBE growth, structural and optical properties of multilayer heterostructures for quantum-cascade lasers

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Abstract. The results obtained in MBE growth and study of the structural and optical properties of GaAs/AlGaAs heterostructures with 228 quantum cascades are presented.

1. Introduction
Sources of the terahertz (THz) frequency range are in demand for fundamental science, commercial interest and special purposes, including spectroscopy, determination of trace amounts of various substances, development of systems forming images of objects concealed from ordinary optical systems, wide-bandwidth communication systems, etc. One of the main problems is the lack of compact coherent sources of radiation of sufficiently high-power for this spectral range. Among various techniques for THz generation [1-3], THz quantum cascade lasers (THz QCL) are compact, coherent, continuous wave (cw) solid-state source with electrical pumping considered to be the most promising THz source. The first QCLs for the THz range were fabricated in the early 2000s [4, 5]. To date THz QCLs of this kind have been developed, which operate up to 200\textdegree K [6] and have an emission power of several tens of mW at liquid-nitrogen temperature. Also, room temperature laser sources of terahertz radiation have been developed [7] based on the intraband amplification of the differential frequency of two QCLs of the mid-infrared (IR) spectral range [8].

In this study, we have investigated the possibility of MBE synthesis of multi-period multilayer heterostructures for use in the fabrication of QCLs for the frequency range around 3 THz.

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We have perform the MBE growth of two multilayer GaAs/AlGaAs heterostructures with different GaAs active layer thicknesses, examine their structural and optical properties by the X-ray diffraction method and photoluminescence (PL) spectroscopy.

2. Experiments.
The epitaxial structures QCL-1 and QCL-2 were synthesized by molecular-beam epitaxy using a Riber 21 MBE machine. Growth was performed on semi-insulating GaAs (100) substrates under arsenic-stabilized conditions. Particular attention was given to accurately setting the growth rates and to maintaining their stability during whole the growth run. The growth rates were carefully calibrated on a separate sample directly prior to growth of the laser structures. The GaAs and AlAs growth rates were set at 0.85 and 0.15 monolayers per second (ML/s) respectively for QCL-1 and 0.7 and 0.105 ML/s for QCL-2. Special high-speed shutters were used to obtain high quality interfaces. In our case, the actuation time of the shutters of the aluminium and gallium sources did not exceed 0.15 s. A 200-nm-thick $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$ stop-layer was deposited onto a GaAs buffer layer. The active region contained 228 periods. Each cascade contains a GaAs/AlGaAs double quantum well (QW), between whose levels a laser transition occurs, and a wider QW serving as the injector/extractor of electrons. The active region was bound from above and below by GaAs:Si contact layers ($5 \times 10^{18} \text{ cm}^{-3}$) with thicknesses of 75 and 50 nm, respectively. The middle part of the injector/extractor layers also had n-type doping to a concentration of $\sim 5 \times 10^{16} \text{ cm}^{-3}$. When heterostructures intended for the generation of light in the THz range are synthesized by the MBE method with total thicknesses of $\sim 10 \mu\text{m}$, the GaAs growth rate may, nevertheless, decrease due to exhaustion of the gallium source in the course of prolonged deposition. Figure 1 shows the schematic diagram of the grown structure. The same circumstance may lead to irreproducibility of the layer thicknesses from one structure to another. In order to elucidate the influence exerted by accurate maintenance of the gallium-arsenide deposition rate, we synthesized and studied two structures, QCL-1 and QCL-2, designed in such a way that the thicknesses of all GaAs layers in QCL-1 are 15% smaller than those of the corresponding layers in QCL-2.

![Figure 1. The schematic diagram of the QCL-1.](image_url)

3. Results and Discussion.
The structural properties of the epitaxial samples were examined by the high-resolution X-ray diffraction (HR-XRD) method with a D8 DISCOVER Bruker AXS diffractometer (radiation wavelength $\lambda =0.15406 \text{ nm}$) with a primary beam half-width of $<12$ arcsec in the $\Omega$–2$\theta$ scanning mode. Figure 2 shows the rocking curve around the symmetric GaAs (004) reflection, measured for the QCL-1 structure. The full width at half-maximum (FWHM) of the satellite peaks due to the periodic repetition of QCL cascades is 15–19 arcsec. It is noteworthy that the full width of the superstructure peaks in the model spectrum, found with allowance for bending of the structure under
elastic stresses, is 22.4 arcsec. This means that both the possible effect of inaccurately maintaining the cascade thickness within the whole structure and the roughness of the heterointerfaces can be neglected, which confirms that the technological parameters for the case of synthesis of the active region are chosen correctly. The calculated rocking curve for the model structure providing the best agreement with the experimental data is also shown in Figure 1. The thickness of the cascade, determined by simulation of the X-ray rocking curves, was 38.3 and 43.6 nm for, respectively, the QCL-1 and QCL-2 structures, instead of the expected values of 38.95 and 43.91 nm. Thus, the discrepancy between the expected and experimentally measured values of the period was 1.7 and 0.7% for the QCL-1 and QCL-2 structures, respectively.

![Figure 2. X-ray rocking curve of the QCL-1 structure near the GaAs (004) reflection and the simulated curve.](image)

In near- and mid-IR lasers operating at interband optical transitions, the lasing wavelength is found to be close to the PL wavelength of the active region. The use of rapid luminescence diagnostics facilities that can be applied to epitaxial structures before performing processes associated with device fabrication strongly simplifies the development of a technology for the fabrication of lasers of this kind, with the design of the laser structure modified when necessary. In the case of THz QCLs, however, there are no direct methods of this kind for rapidly determining the expected lasing wavelengths. In this context, it seems to be effective to compare the results obtained in measuring the energies of electron–hole optical transitions and correlate these energies with calculation results. In the case of their good agreement, it can be assumed that the energies of the intraband transitions between electron levels of the cascades will also coincide with those predicted by simulation results.

The optical properties were studied by PL spectroscopy. Measurements were made in the temperature range T = 77–300°K. Optical pumping was provided by a YAG:Nd laser operating at the second harmonic in the continuous-wave mode (wavelength λ = 527 nm), with the pump power density varied from 2.5 to 15 W/cm². The PL signal was detected with an FHR 1000 monochromator and a single-channel cooled Si photodiode.
Figure 3. (a) Band diagram of a biased QCL-2 structure and (b) relationship between the oscillator strength and transition frequency for QCL-1 and QCL-2.

Figure 3a shows the PL spectra of the QCL-1 structure, measured at 77 K and various excitation densities in the range $P = 2.5–15$ W/cm$^2$, and Figure 3b, spectra of the QCL-2 structure, measured at a fixed power density (15 W/cm$^2$) and various temperatures in the range $T = 77–140$°K. Three emission peaks can be distinguished in the spectra of both structures, identified as being associated with transitions between the first electron level and the first level of heavy holes (E1–HH1), the first electron level and the first level of light holes (E1–LH1), and the second electron level and the second level of heavy holes (E2–HH2). This interpretation of the peaks agrees with the fact that the relative intensity of shorter wavelength peaks increases with increasing excitation level, and when the temperature changes, the spectral position of the peaks moves in accordance with the temperature behavior of the band gap of GaAs. It is noteworthy that high-intensity luminescence at the wavelength of the ground optical transition E1–HH1 was observed in both structures up to room temperature.

Thus, the method of molecular-beam epitaxy was used to synthesize multiperiod (228 cascades, $\sim$10 μm) GaAs/AlGaAs epitaxial structures intended for use in the fabrication of quantum-cascade lasers for the terahertz range. The experimental value of the cascade thickness corresponds to the expected value with an accuracy of better than 2%. The angular width of the superstructure peaks in the X-ray rocking curve does not exceed 20 arcsec. A study of the photoluminescence spectra demonstrated good agreement (within 1 meV) between the interband optical-transition energies and the calculated values. A 15% change in the thickness of the GaAs layers in two different structures leads to expected shifts of the positions of the optical transitions. This means that the positions of energy levels in complex structures of this kind can be estimated by various express optical diagnostic procedures operating in the near-IR range.

The technology for the fabrication of waveguides of the metal–semiconductor–metal type is being developed in order to fabricate laser diodes from the structures has been investigated this report. Finally, the laser-related I-V curves were recorded at 10°K.

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