Technical Physics Letters, 2022, Vol. 48, No. 5

11.3

3.8 THz quantum cascade laser grown by metalorganic vapor phase epitaxy

© T.A. Bagaev[1], M.A. Ladugin[1], A.A. Marmalyuk[1], A.I. Danilov[1], D.V. Ushakov[2], A.A. Afonenko[2], A.A. Zaytsev[3], K.V. Maremyanin[4], S.V. Morozov[4], V.I. Gavrilenko[4,5], R.R. Galiev[6], A.Yu. Pavlov[6], S.S. Pushkarev[6], D.S. Ponomarev[6], R.A. Khabibullin[5,6]

[1] Polyus' Research Institute of M.F. Stelmakh Joint Stock Company, Moscow, Russia
[2] Belarusian State University, Minsk, Republic of Belarus
[3] National Research University of Electronic Technology, Zelenograd, Moscow, Russia
[4] Institute of Physics of Microstructures, Russian Academy of Sciences, Nizhny Novgorod, Russia
[5] Ioffe Institute, St. Petersburg, Russia
[6] V.G. Mokerov Institute of Ultra High Frequency Semiconductor Electronics of RAS, Moscow, Russia
E-mail: khabibullin@isvch.ru

Received February 11, 2022
Revised March 14, 2022
Accepted March 22, 2022

We have demonstrated a quantum cascade laser (QCL) with a generation frequency of about 3.8 THz, grown by metal-organic vapor phase epitaxy. The multilayer heterostructure for QCLs consists of 185 repetitions of an active module containing four GaAs/$Al_{0.15}Ga_{0.85}As$ quantum wells. The threshold current and threshold voltage of the fabricated QCL were 2.25 kA/cm^2 and 19.7 V, respectively. The QCL oscillations were carried out in the multimode regime, and the detection of terahertz radiation continued with an increase in the laser temperature up to 60 K.

Keywords: quantum cascade lasers, terahertz, metalorganic vapor phase epitaxy, quantum well, QCL, MOCVD, MOVPE.

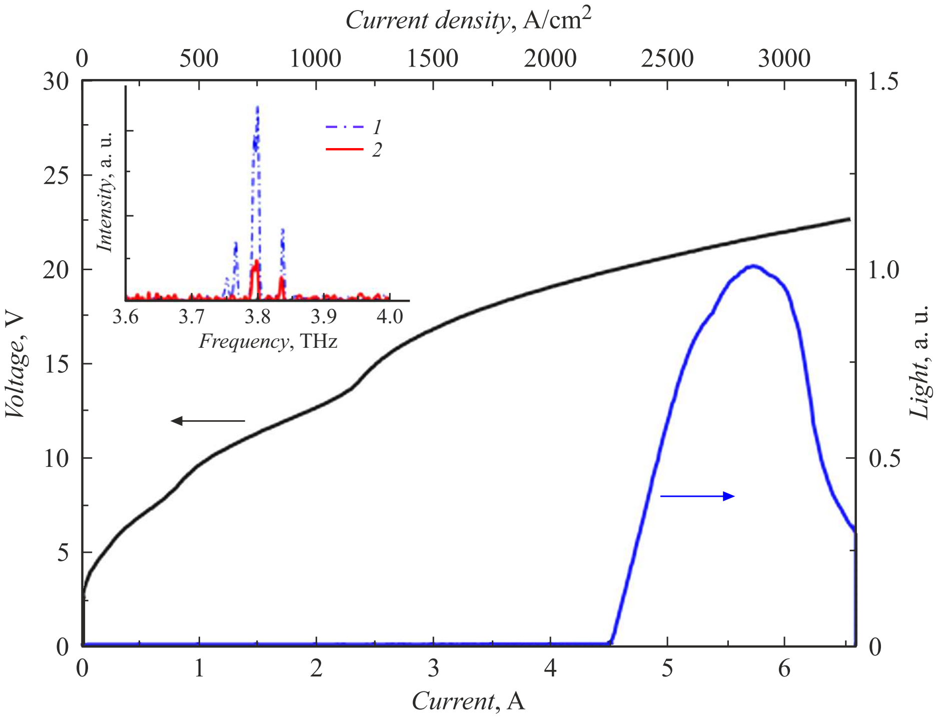
DOI: 10.21883/TPL.2022.05.53479.19162

The extremely high requirements for epitaxial growth of multilayer heterostructures for terahertz quantum cascade lasers (THz QCL) result in high costs and an extremely limited supply of these lasers on the market. The low optical gain of the intersubband transitions and high loss in the resonator forces a „cascading scheme" with an active region thickness of about 10 μm, which requires multiple (over 100 times) repetitions of the active module — period of the structure consisting of a set of quantum wells (QW) [1,2]. The growth times of such structures exceed 10 hours, therefore, the stability of growth rates of semiconductor layers over long periods of time is a matter of concern.

The vast majority of successful THz QCLs are based on GaAs/AlGaAs structures grown by molecular beam epitaxy (MBE) [3]. The MBE technology makes it possible to grow structures with sharp heterojunctions and low levels of background doping. The development of MBE growth technology and THz QCL band designs over the last 20 years has enabled high operating temperatures of lasers up to 250 K (about -23°C) with the possibility of using thermoelectric cooling on a Peltier element [4]. However, the stability of the growth rate in MBE is determined by the temperature maintenance level of the effusion cells with III group materials (Ga and Al), which cannot be in thermodynamic equilibrium with the crucible during growth because of the shutter operation. It has previously been shown that using *in situ* techniques to stabilize the growth rate in MBE can significantly improve the repeatability of layer thicknesses and composition both from period to period in a single structure as well as in a series of grown structures, which improves the performance of manufactured THz QCLs [5].

The growth of semiconductor structures by metalorganic vapor phase epitaxy (MOVPE) has traditionally been regarded as inferior to MBE in terms of the heterojunctions sharpness and the concentration of background impurities. However, the use of the MOVPE growth mode at low pressure in a reactor with lower growth rates also produces sharp boundaries of heterojunctions. The stability of the growth rate in MOVPE is determined by the accuracy of maintaining the growth temperature and gas pressure, which is, at first glance, technically easier than stabilizing the MBE growth rate. In addition, the use of industry-oriented MOVPE technology with the ability to scale the reactor to grow more structures at the same time has a cost advantage in terms of a single structure.

To date, the successful implementation of THz QCLs based on GaAs/AlGaAs grown by MOVPE has only been demonstrated by one research group led by E. Kapon from EPFL (Switzerland), as reported in two papers [6,7]. These papers show that THz QCLs on MOVPE and MBE structures exhibit close values of maximum operating temperature $T_{\max}$. Another successful example is InGaAs/InAlAs-based QCLs with generation frequency 3.7 THz grown by MOVPE method, which had $T_{\max}$ higher than that of identical in zone design QCLs based on MBE structures [8]. Thus,



Current-voltage characteristic and current dependence of integral emission intensity for THz QCLs measured at 4.2 K in pulsed mode with a pulse duration of 1 μs and a repetition rate of 100 Hz. The inset shows the laser generation spectra measured in pulsed mode at about 6 A current and two temperatures: 12 (*1*) and 60 K (*2*).

the use of MOVPE technology for growth of GaAs/AlGaAs-based THz QCLs needs to be researched in more detail.

The aim of this paper is to demonstrate the feasibility of making THz QCLs with an active module of four GaAs/Al$_{0.15}$Ga$_{0.85}$As QWs based on a multilayer heterostructure grown by MOVPE.

To design the GaAs/Al$_{0.15}$Ga$_{0.85}$As heterostructure, a five-level laser scheme with resonant-phonon emission of the lower laser level and an optical gain maximum of about 3.8 THz was chosen for QCL. Injection of electrons into the upper laser level is achieved using a resonant tunneling mechanism. The total thickness of the QWs and barriers of one active module (structure period) is about 55 nm, which requires 185 periods growth to achieve an active region thickness of about 10 μm. The central part of the wide QW of the active module needs to be doped with a donor impurity with a layer concentration of $4.6 \cdot 10^{10}\,\mathrm{cm}^{-2}$. The concentration of donor doping in the contact layers is required at $5 \cdot 10^{18}\,\mathrm{cm}^{-3}$ to minimize the parasitic resistance of the metal/semiconductor contact. The double metal waveguide requires the formation of an AlGaAs stop-layer between the substrate and the heterostructure with a thickness of more than 200 nm with an aluminum content of more than 55% to ensure the selectivity of wet etching.

The GaAs/AlGaAs multilayer heterostructure for QCLs was grown by MOVPE at low pressure in a horizontal reactor at POLYUS Research Institute of M.F. Stelmakh. For epitaxial growth, triethyl gallium, trimethylaluminum and arsine were used as precursors of Ga, Al and As. The growth rate of GaAs layers was 0.180 nm/s and AlGaAs — 0.214 nm/s when the active region of QCL consisting of Al$_{0.15}$Ga$_{0.85}$As barriers and GaAs quantum wells with minimum thickness of 8 and 30 monolayers, respectively. The accuracy of semiconductor superlattice formation, including the stability of layer parameters from the first to the last period, is important for the efficient generation of THz QCL. To this end, this paper uses approaches to obtain sharp hetero-boundaries and control the growth rate successfully tested for growing AlGaAs/GaAs heterostructures with ultra-thin layers and a large number of periods [9].

A square-shaped sample of approximately 1 × 1 cm was out of the grown MOVPE structure, from which the double metal waveguide based on gold were manufactured. The manufacturing method is cleaved in detail in [10,11]. Unburned Ti/Au contacts were used for the electrical contact to the laser structure, resulting in an additional voltage drop (3–4 V) across the Schottky barrier. Cleaved THz QCLs with Fabry–Perot resonator (width 100 μm, length 2 mm) were mounted on a C-mount type heat sink; electrical contact was made by welding several 30 μm diameter gold wires, distributed evenly over the entire length of the laser strip.

The figure shows the current-voltage characteristic and the dependence of the integral signal on the injection current for the studied THz QCL (No. 33111), measured at a temperature of 4.2 K in pulsed mode with a pulse duration of 1 μs and a repetition frequency of 100 Hz. A more detailed description of the measurement method is given in [12]. It should be noted that in comparison to [6], where the authors faced the problem of doping the contact layers during MOVPE growth and obtained large QCL operating voltages (over 40 V), in our case, the QCL operating voltages of about 20 V correspond well to the calculated values of external electric field which should be applied to the structure for optimum electron level alignment. The large threshold generation current of the manufactured QCL (about 4.5 A, corresponding current density 2.25 kA/cm^2) is characteristic of QCLs with resonance-phonon designs, further exacerbated by the use of a five-level laser scheme. At the same time, the large dynamic range of generation currents, ranging from 4.5 to 7.5 A, should be noted. The inset to the figure shows the spectra of QCL generation at a current of about 6 A, measured with a Fourier spectrometer at temperatures of 12 and 60 K. The highest intensity is in the radiation mode with a frequency of about 3.8 THz, which agrees well with the calculated gain spectrum for the chosen design. At temperatures around 60 K the emission intensity of the QCL was still high enough to measure the spectrum. Taking into account the unavoidable loss of terahertz radiation when measuring the generation spectra, we assume that the maximum operating temperature of the manufactured QCL should be at least 70 K.

Thus, this paper has successfully demonstrated the possibility of MOVPE growth of GaAs/AlGaAs multilayer heterostructure for THz QCL with an active region thickness of about 10 μm. The THz MOVPE-based QCL emits near 3.8 THz while retaining sufficient intensity to measure the spectrum when the temperature is increased to 60 K. We believe that continued work on MOVPE growth of THz QCL will improve the performance of lasers potentially capable of competing with THz QCL based on MBE structures.

Acknowledgments

The authors would like to thank A.S. Bugaev V.G. Mokerov Institute of Ultra High Frequency Semiconductor Electronics of RAS for valuable discussions on epitaxial techniques.

Funding

The work on the post-strength fabrication and measurement of QCL radiation characteristics was supported financially by the Russian Science Foundation (Grant № 21-72-30020, https://rscf.ru/en/project/21-72-30020/).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] A.E. Yachmenev, S.S. Pushkarev, R.R. Reznik, R.A. Khabibullin, D.S. Ponomarev, Prog. Cryst. Growth Charact. Mater., **66**, 100485 (2020). DOI: 10.1016/j.pcrysgrow.2020.100485

[2] D.V. Ushakov, A.A. Afonenko, A.A. Dubinov, V.I. Gavrilenko, I.S. Vasil'evskii, N.V. Shchavruk, D.S. Ponomarev, R.A. Khabibullin, Quantum Electron., **48** (11), 1005 (2018). DOI: 10.1070/QEL16806.

[3] M.S. Vitiello, A. Tredicucci, Adv. Phys. X, **6**, 1893809 (2021). DOI: 10.1080/23746149.2021.1893809

[4] B. Wen, D. Ban, Prog. Quantum Electron., **80**, 100363 (2021). DOI: 10.1016/j.pquantelec.2021.100363

[5] L.H. Li, J.X. Zhu, L. Chen, A.G. Davies, E.H. Linfield, Opt. Express, **23**, 2720 (2015). DOI: 10.1364/OE.23.002720

[6] L. Sirigu, A. Rudra, E. Kapon, M.I. Amanti, G. Scalari, J. Faist, Appl. Phys. Lett., **92**, 181111 (2008). DOI: 10.1063/1.2924294

[7] M.I. Amanti, G. Scalari, R. Terazzi, M. Fischer, M. Beck, J. Faist, A. Rudra, P. Gallo, E. Kapon, New J. Phys., **11**, 125022 (2009). DOI: 10.1088/1367-2630/11/12/125022

[8] K. Fujita, M. Yamanishi, S. Furuta, K. Tanaka, T. Edamura, T. Kubis, G. Klimeck, Opt. Express, **20**, 20647 (2012). DOI: 10.1364/OE.20.020647

[9] M.A. Ladugin, I.V. Yarotskaya, T.A. Bagaev, K.Yu. Telegin, A.Yu. Andreev, I.I. Zasavitskii, A.A. Padalitsa, A.A. Marmalyuk, Crystals, **9**, 305 (2019). DOI: 10.3390/cryst9060305

[10] R.A. Khabibullin, N.V. Shchavruk, D.S. Ponomarev, D.V. Ushakov, A.A. Afonenko, I.S. Vasil'evskii, A.A. Zaycev, A.I. Danilov, O.Yu. Volkov, V.V. Pavlovskiy, K.V. Maremyanin, V.I. Gavrilenko, Semiconductors, **52** (11), 1380 (2018). DOI: 10.1134/S1063782618110118.

[11] R.A. Khabibullin, N.V. Shchavruk, D.S. Ponomarev, D.V. Ushakov, A.A. Afonenko, K.V. Maremyanin, O.Yu. Volkov, V.V. Pavlovskiy, A.A. Dubinov, Opto-Electron. Rev., **27**, 329 (2019). DOI: 10.1016/j.opelre.2019.11.002

[12] A.V. Ikonnikov, K.V. Marem'yanin, S.V. Morozov, V.I. Gavrilenko, A.Yu. Pavlov, N.V. Shchavruk, R.A. Khabibullin, R.R. Reznik, G.E. Cirlin, F.I. Zubov, A.E. Zhukov, Zh.I. Alferov, Tech. Phys. Lett., **43** (4), 362 (2017). DOI: 10.1134/S1063785017040083.