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RESEARCH ARTICLE

High-temperature terahertz quantum-cascade lasers: design optimization and experimental results

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Abstract
Objectives. Terahertz quantum-cascade lasers (THz QCLs) are compact solid-state lasers pumped by electrical injection to generate radiation in the range from 1.2 to 5.4 THz. The THz QCL operating frequency band contains absorption lines for a number of substances that are suitable for biomedical and environmental applications. In order to reduce the size and cost of THz QCLs and simplify the use of THz sources in these applications, it is necessary to increase the operating temperature of lasers.
Methods. To calculate electron transport in THz QCLs, we used a system of balance equations based on wave functions with reduced dipole moments of tunnel-bound states.
Results. As a result of the calculations, an original band design with a period based on three $GaAs/Al_{0.18}Ga_{0.82}As$ quantum wells (QWs) and a gain maximum at about 3.3 THz was proposed. Based on the developed design, a THz QCL was fabricated, including the growth of a laser structure by molecular beam epitaxy, postgrowth processing to form strip lasers with a double metal waveguide, as well as an assembly of lasers mounted on a heat sink. The developed THz QCLs was capable of lasing at temperatures of up to 125 K as predicted by the performed calculations. We also studied band designs based on two $GaAs/Al_xGa_{1-x}As$ QWs having varying aluminum contents in the barrier layers (x = 0.20, 0.25, and 0.30).
Conclusions. The calculated temperature dependences of the peak gain for two-QW designs with $x > 0.2$ confirm the possibility of creating THz QCLs operating at temperatures above 200 K. Thus, we have proposed two-QW band designs that outperform existing high-temperature designs in terms of maximum operating temperature.

Keywords: quantum cascade laser, terahertz range, quantum well, molecular beam epitaxy

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НАУЧНАЯ СТАТЬЯ

Высокотемпературные квантово-каскадные лазеры терагерцового диапазона: оптимизация дизайнов и экспериментальные результаты

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Резюме
Цели. Квантово-каскадные лазеры терагерцового диапазона (ТГц ККЛ) являются компактными твердотельными приборами с инжекционной накачкой, которые позволяют генерировать излучение в диапазоне от 1.2 до 5.4 ТГц. В полосе рабочих частот ТГц ККЛ находятся линии поглощения для ряда веществ, актуальных для медико-биологических и экологических приложений. Для широкого применения ТГц ККЛ в данных приложениях необходимо увеличивать рабочую температуру лазеров, что позволит уменьшить размеры и стоимость ТГц ККЛ, а также упростит использование данных ТГц-источников.
Методы. В работе для расчета электронного транспорта в ТГц ККЛ использовалась система балансных уравнений на основе базиса волновых функций с уменьшенными дипольными моментами туннельно-связанных состояний.
Результаты. В результате расчетов предложен оригинальный зонный дизайн с периодом на основе трех $GaAs/Al_{0.18}Ga_{0.82}As$ квантовых ям (КЯ) и максимумом усиления около 3.3 ТГц. На основе разработанного дизайна был экспериментально изготовлен ТГц ККЛ, что включало рост лазерной структуры методом молекулярно-лучевой эпитаксии, постростовой процессинг для формирования полосковых лазеров с двойным металлическим волноводом и сборку лазеров на теплоотводе. Изготовленные ТГц ККЛ продемонстрировали генерацию вплоть до температуры 125 K, что согласуется с проведенными расчетами. Также в работе проведено исследование зонных дизайнов на основе двух $GaAs/Al_xGa_{1-x}As$ КЯ с различным содержанием алюминия в барьерных слоях (x = 0.20, 0.25 и 0.30).
Выводы. Рассчитанные температурные зависимости пикового усиления для двух-КЯ дизайнов с $x > 0.2$ подтверждают возможность создания ТГц ККЛ, работающих при температурах свыше 200 K. Таким образом, в работе предложены двух-КЯ зонные дизайны, которые превосходят по максимальной рабочей температуре существующие рекордные высокотемпературные дизайны ТГц ККЛ.

Ключевые слова: квантово-каскадный лазер, терагерцовый диапазон, квантовая яма, молекулярно-лучевая эпитаксия

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INTRODUCTION

Both in terms of fundamental science and as comprising a potential solution to a large number of applied problems, the terahertz frequency range (1–10 THz) remains one of the most intriguing regions of the electromagnetic spectrum. For a long time, the lack of compact solid-state sources of THz radiation with an average power level measured in mW remained a limiting factor for realizing the advantages of THz waves in spectroscopy, visualization, and remote sensing applications. The possibility of "transferring" the operational scheme of quantum-cascade lasers (QCLs) in the mid-infrared (mid-IR) range to the THz region demonstrated the possibility of tuning the generation frequency from 1.2 to 5.4 THz to create a unique source of THz radiation by changing the thickness of the semiconductor layers without using large magnetic fields. However, in comparison with mid-IR QCLs, where operating temperatures of more than 300 K have been demonstrated, the operation of THz QCLs required long periods of cryogenic cooling. Thus, the task of raising THz QCL operating temperatures using thermoelectric cooling based on the Peltier effect becomes highly relevant in order to for THz QCLs to find practical application in medicine, biology, agriculture, ecology, the fight against terrorism, and wireless communication [1–8].

The cryogenic cooling used with the first THz QCLs, which required the laser chip to be mounted on the cold finger of a flooded cryostat or a closed cycle cryocooler, significantly affected the size and energy efficiency of such THz sources, complicating the use of this type of laser outside laboratories. The optimization of THz QCL band designs and use of low-loss waveguides led to an increase in the operating temperature of these lasers, allowing the use of compact electric cryocoolers operating according to the Stirling cycle, which do not require cryogenic liquids and have a service life of more than 30 000 hours. However, the main disadvantage of this approach is the high cost of Stirling cryocoolers (tens of thousands of US dollars), which also limited the widespread use of THz QCLs.

Studies into electron transport [9] and the creation of more efficient band designs of the THz QCL active region [10] led to the revolutionary demonstration in 2019 of the first thermoelectrically cooled lasers operating at a maximum temperature of 210.5 K (−63°C). By the end of 2020, data had been published demonstrating a THz QCL having a maximum operating temperature of 250 K (about −23°C) [11], which permitted the use for cooling of a single-stage Peltier element costing less than USD 100. Meanwhile, research featuring similar band designs whose active region period was based on two quantum wells (QWs), differing in the height of potential barriers $Al_xGa_{1-x}As_x$ = 0.25 ([10], 2019) and $x = 0.30$ ([11], 2020), was published. Here, the band design and approach to increasing the height of potential barriers are primarily aimed at suppressing parasitic channels of electron leakage into the continuum, since the presence of such parasitic conduction channels in the active region adversely affects the operation of THz QCLs at high temperatures [12]. The increase in the height of potential barriers results in the need to reduce their thickness in order to maintain tunneling transparency, which in turn leads to even higher requirements for the epitaxial growth of such heterostructures having extremely thin barrier layers with a thickness of a few monolayers [13].

Despite work on the creation of THz QCLs in Russia having begun with a 10–15-year delay relative to the work of foreign groups, THz QCLs exclusively fabricated in Russia include the use of molecular beam epitaxy (MBE) to grow heterostructures and post-growth processing of stripe lasers with a double metal waveguide [14, 15]. In this paper, we present the experimental results of a study into THz QCLs based on an original active region design whose period contains three $GaAs/Al_{0.18}Ga_{0.82}As$ QWs. The results of the band design optimization of THz QCLs having a period based on two $GaAs/Al_xGa_{1-x}As$ QWs with x = 0.20, 0.25, and 0.30 for operation at high temperatures (greater than 200 K) are also presented.

EXPERIMENTAL STUDY OF THE THz QCL WITH A PERIOD BASED ON THREE $GaAs/Al_{0.18}Ga_{0.82}As$ QWs

The highest THz QCL operating temperatures to date have been demonstrated by a resonant phonon (RP) design featuring successive resonant tunneling of electrons to the upper laser level and emission of a THz photon, along with the depopulation of the lower laser level due to electron scattering on longitudinal optical phonons (longitudinal-optical phonons, LO-phonons). By means of diagonal radiative transitions, it was possible to significantly reduce the rate of nonradiative recombination of "hot" electrons from upper to lower laser levels due to the temperature activation of the emission of LO-phonons. Based on the foregoing, in order to optimize the active region of a high-temperature THz QCL, we selected a resonant-phonon design having a period of three $GaAs/Al_{0.18}Ga_{0.82}As$ QWs and a generation frequency of about 3.3 THz.

In order to achieve the maximum operating temperature, numerical optimization of the design of the THz QCL active region was carried out according to the previously developed method based on a system of balance equations [16, 17]. In the course of optimization, the thicknesses of all layers of the period were scanned over a wide range (thicknesses of the QWs and barriers) with a step equal to half the constant of the GaAs crystal lattice. When selecting optimal designs, there was a condition for ensuring maximum gain on the positive branch of the current-voltage characteristic in order to avoid electrical instabilities that prevent the formation of electric field domains. The maximum operating temperature of the design corresponded to the intersection point of the temperature dependences of the gain and losses in the resonator, which were calculated earlier for a THz QCL with a double metal waveguide [16].

In the course of the optimization, an original $GaAs/Al_{0.18}Ga_{0.82}As$ band design was developed based on a period comprising a sequence of layer thicknesses starting from the injector barrier: 4.23/**16.09**/3.95/**8.75**/2.54/**8.18** (in units of nm, GaAs QWs are in bold). Doping of the active module with a Si donor impurity was $3.0 \cdot 10^{10}$ cm^{-2}. In the model, the height of $Al_{0.18}Ga_{0.82}As$ potential barriers in the conduction band was assumed to be ΔE_c = 164 meV.

An illustration of electronic transport in the developed band design is shown in Fig. 1; here, the current flow through the electron levels in two periods is indicated by arrows. The number of arrows between levels is proportional to the current density (the current density through the period corresponds to five arrows). During optimization, spurious conduction channels having current densities of less than one arrow were minimized (not shown in Fig. 1). The diagonal laser transition E_{43} with a matrix element of dipole transitions Z_{43} = 3.8 nm corresponds to a generation frequency of 3.3–3.4 THz. In the insert, Fig. 1 shows the gain spectrum of the developed design (red line) and the loss spectrum in the resonator (blue dot-dash line).

A laser structure based on the developed $GaAs/Al_{0.18}Ga_{0.82}As$ band design was grown by MBE

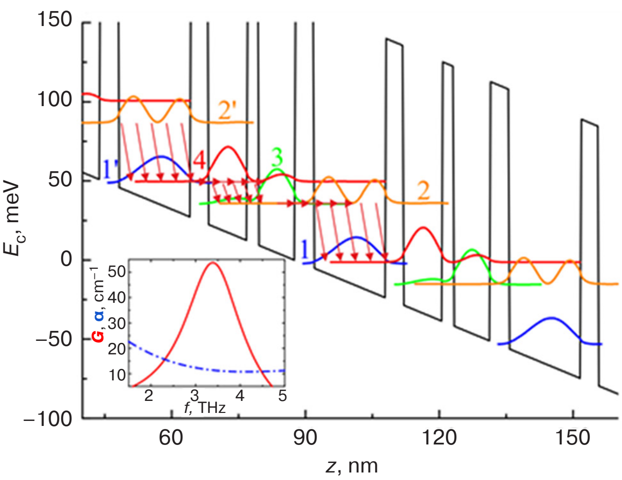

Fig. 1. Energy profile of the bottom of the conduction band E_c; energy levels (numbered) and basis wave functions after the localization procedure (tight coupling model) for two successive active modules at voltage V_1 = 51 mV and temperature 77 K

on a Riber Compact 21T setup at the National Research Nuclear University "MEPhI" (group of Prof. I.S. Vasilevsky). When developing optimal growth parameters, special attention was paid to the selection of technological growth conditions (substrate temperature, As_4 arsenic flow), accurate calibration of the growth rates of GaAs and AlAs compounds, as well as their stability during the growth of thick structures. Excluding the time taken for the growth of gauge superlattices, which were grown before and after the growth of the laser structure, the growth time of one laser structure was more than 10 h. The deviation of the thickness of the period of the structure grown from the nominal thickness (specified in the design) did not exceed 1%.

Based on the grown laser structure, THz QCLs with a double metal waveguide (DMW) were fabricated. The fabrication procedure for stripe lasers with Au–Au DMW is described in detail in [18, 19]. The cleaved THz QCLs with a Fabry–Perot resonator were assembled at the M.F. Stelmakh Polyus Research Institute (group of S.M. Sapozhnikov) on C-mount radiators; electrical contact was made by welding to the upper metal of the laser strip a large number of gold wires with a diameter of 30 μm, distributed evenly along the entire length of the strip to improve electrical injection over the entire surface of the laser [20].

The radiation characteristics of the fabricated lasers were measured at the Institute of Physics of Microstructures of the Russian Academy of Sciences (group of Prof. V.I. Gavrilenko). The current-voltage characteristics (*I–V*) and the dependences of the integral radiation intensity on the pump current and temperature of the fabricated THz lasers were studied in a pulsed mode (pulse duration 500 ns, repetition frequency 100 Hz). The structures were powered by a specially made electronic key, which made it possible to obtain pulses of a given duration, duty cycle, and amplitude, as well as to measure the voltage and current passing through the laser. When measuring the voltage-current characteristics and the integral dependences of radiation on current/temperature, the signal from the detector was fed to a Stanford Research Systems SR250 two-channel strobe integrator (Sunnyvale, California, USA).

In order to determine the maximum operating temperature, we measured the dependences of the integrated radiation intensity on the pump current. Figure 2 shows that an increase in temperature from 8 to 60 K has little effect on the output power or threshold current values. When approaching 100 K, the intensity of the optical signal begins to decrease sharply. The rapid increase in the threshold current at temperatures above 100 K is most likely due to the activation of parasitic conduction channels. The maximum temperature at which lasing was observed was about 125–130 K. A metal tube having its own loss level in the THz range was used as a waveguide system for extracting THz QCL radiation from the cryostat. It can be assumed that the actual maximum operating temperature of the THz QCL is higher by 5–10 K; that is, about 140 K.

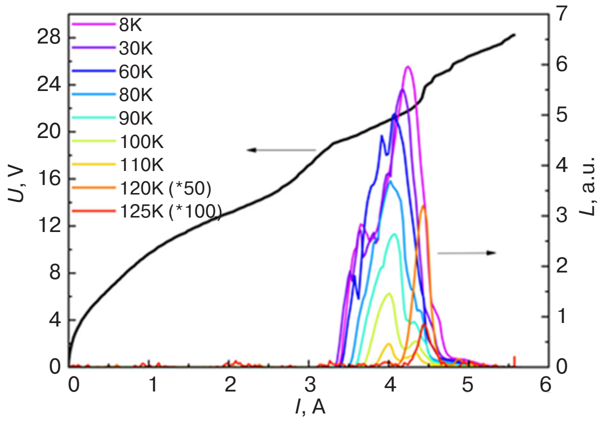

Fig. 2. *I–V* (black line) and current dependences of the integrated radiation intensity for QCL #52111 in the temperature range from 8 to 125 K. The amplitude of the integrated signal for temperatures of 120 and 125 K is multiplied by 50 and 100, respectively

OPTIMIZATION OF THz QCL BAND DESIGN BASED ON TWO GaAs/Al_xGa_{1-x}As QUANTUM WELLS WITH x = 0.20, 0.25, AND 0.30

To further improve the high-temperature characteristics of THz QCLs, it is necessary to develop new concepts for the band design of active regions for reducing losses in the resonator. Approaches for reducing losses in THz QCLs proposed earlier include those based on the use of a double silver metal waveguide [21, 22]. In order to increase the maximum operating temperature of THz QCLs, we proposed to study band designs based on two GaAs/Al_xGa_{1-x}As QWs with different potential barrier heights: x = 0.20, 0.25, and 0.30.

A system of balance equations with a basis of wave functions in the tight-coupling approximation was used to calculate the THz QCL characteristics. This basis was obtained as a superposition of the eigenstates of the Schrödinger equation for the entire THz QCL active region by minimizing the spatial expansion of the wave functions of the tunnel-coupled states. Due to the small wave function overlap (and accordingly low scattering rate) of degenerate ground states with $\Delta E \lesssim 3$ meV, the localized basis of wave functions in the tight-coupling approximation is more resistant to the influence of the dephasing effect. After analyzing band designs of THz QCLs based on two GaAs/Al_xGa_{1-x}As QWs, we optimized the height of potential barriers by varying the aluminum contents (see Table).

The depiction of the proposed designs in Fig. 3 shows the current through the energy levels (red arrows). Several conduction channels can be seen between the electronic levels, including the 3–2 transition between the upper and lower laser levels, as well as parasitic transitions between levels 3 and 1. The temperature dependences of the maximum gain at a frequency of 3.9 THz shown in Fig. 4 demonstrate the possibility of operation at temperatures above 200 K for designs with $x > 0.2$. As a comparison, the calculated dependence of the gain on temperature (blue line) is shown for the band design based on three GaAs/$Al_{0.25}Ga_{0.75}$As QWs proposed in [10], along with the experimental maximum operating temperature T_{max} = 210.5 K.

Table. Parameters of optimized band designs of an active module THZ QCL based on two quantum wells GaAs/Al_xGa_{1-x}As

Design	x	Barrier heigh ΔE_c, meV	Sequences of barriers in a period: **Potential barrier** / quantum well, nm
Bosco [10]	0.25	212.5	**1.98**/16.37/**3.39**/7.91
A	0.20	178.5	**2.3**/16.4/**4.2**/7.3
B	0.25	212.5	**2.0**/17.2/**3.4**/7.9
C	0.30	244.5	**1.7**/17.0/**3.1**/7.9

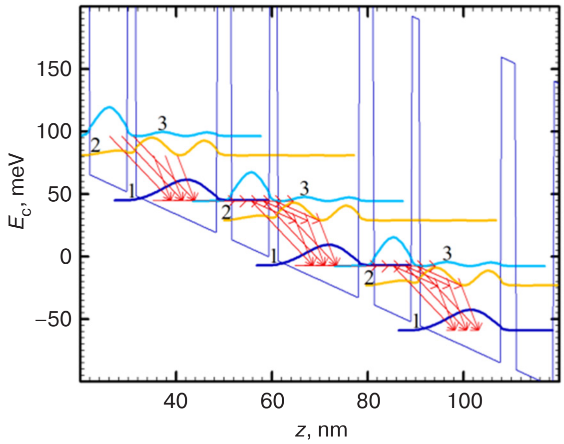

Fig. 3. Band diagram for design C

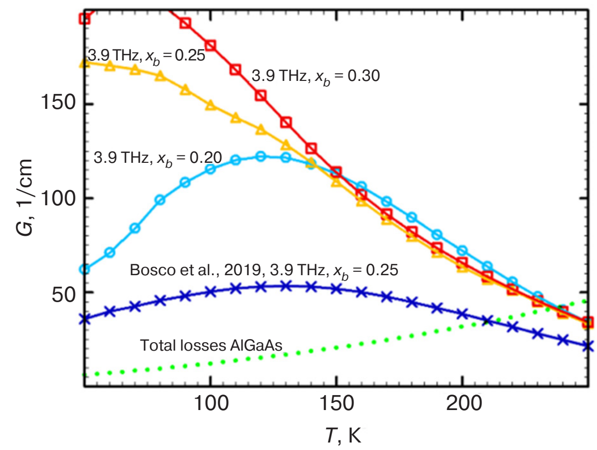

Fig. 4. Temperature dependences of the maximum gain G for the GaAs/Al_xGa_{1-x}As structure with different Al content in the barriers

Designs with gain exceeding losses at high temperatures (210–240 K) were optimized by scanning the thickness of the active module in the range from 24 to 36 nm. In this case, the thickness of the $Al_xGa_{1-x}As$ barrier layers was varied in the range of 1.4–4.0 nm, while that of the GaAs quantum wells was in the range of 6.3–20.0 nm; here, a step was equal to half the lattice constant of GaAs. The parameters of optimized designs of active modules for different compositions of barrier layers (x contents) are presented in Table. In the sequence of period layers, GaAs quantum wells are shown in bold type. The central part of the wide GaAs QW is doped with silicon having a layer concentration of $4.5 \cdot 10^{10}$ cm^{-2}, as in [3].

The main factors affecting the gain in QCLs are the effective electron mass m_e, population inversion n_{32} of laser levels, and matrix elements of dipole transitions Z_{32} [23–25]. Thus, the gain of the QCL active module can be represented as the product of these parameters $G \sim m_c^{-3/2}\ n_{32}\ Z_{32}^2$. This means that effective amplification in a QCL requires a small effective electron mass combined with a large population inversion and dipole transition matrix elements. The temperature dependences of these values for the optimized active modules compared to the Bosco design [3] are shown in Fig. 5, while Fig. 5a depicts the effective electron mass increases in designs with higher potential barriers (higher aluminum content in $Al_xGa_{1-x}As$) due to the non-parabolic effect. For all designs under study, m_c can be seen to decrease at elevated temperatures. In optimized designs B and C (Fig. 5b), the population inversion is larger at temperatures above 200 K. As can be seen from Fig. 5c, the matrix elements of the dipole junctions of these designs increase with rising temperature from 50 to 250 K: from 4.9 to 5.2 nm (design Bosco 2019), from 4.7 to 5.0 nm (design A), from 4.4 to 4.7 nm (design B), and from 4.7 to 5.0 nm (design C). Thus, by combining all three factors for all optimized designs A (x_b = 0.20), B (x_b = 0.25) and C (x_b = 0.30), we can expect a maximum operating temperature of 238 K.

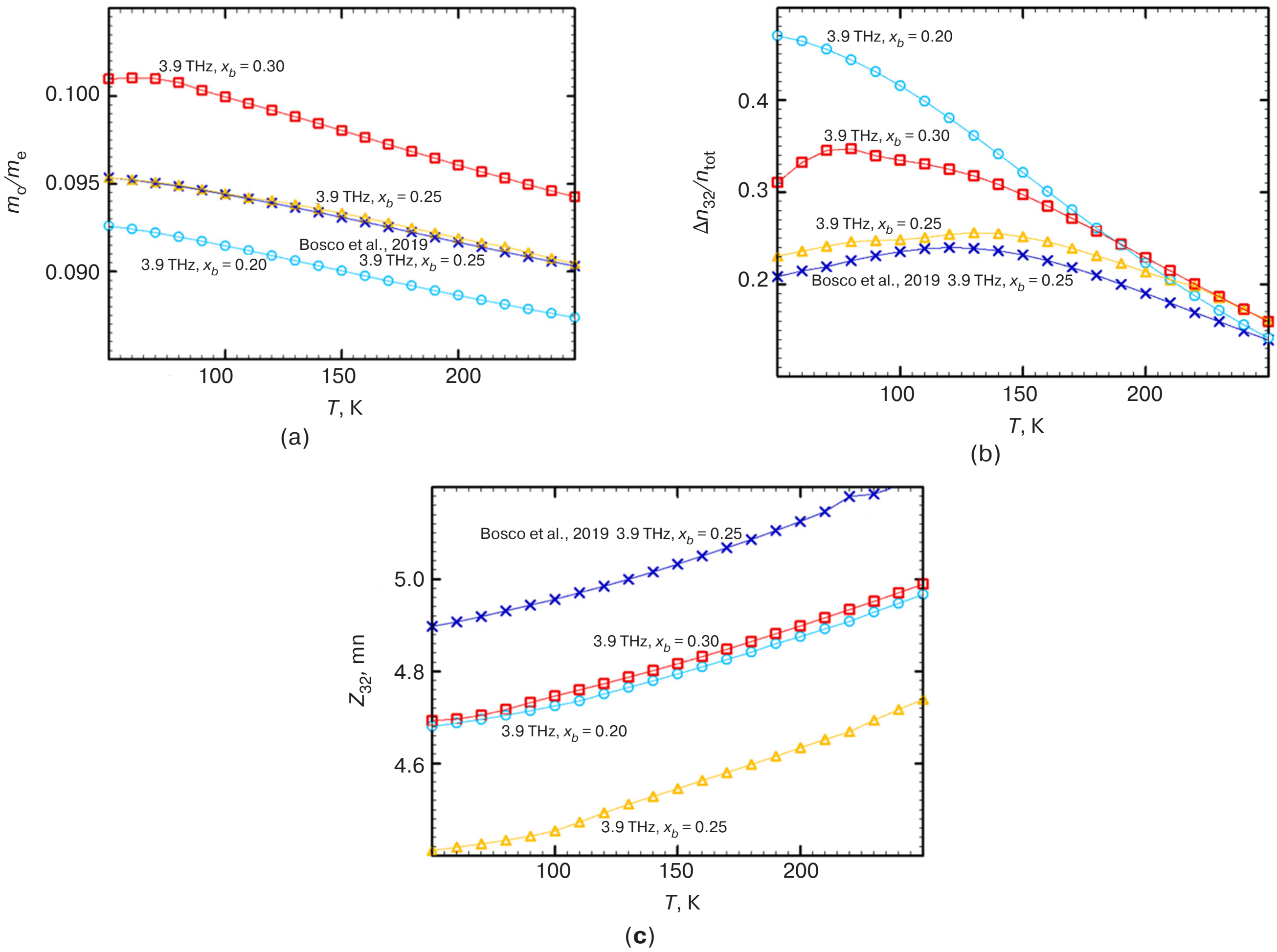

Fig. 5. Temperature dependences of the effective electron mass m_c (a), Δn_{32} are population inversions, n_{tot} is the total electron concentration in a period (b), and matrix elements of dipole transitions Z_{32} (c) for the optimized design A (x_b = 0.20), B (x_b = 0.25), and C (x_b = 0.30)

CONCLUSIONS

A THz QCL based on an original band design having a period based on three $GaAs/Al_{0.18}Ga_{0.82}As$ quantum wells has been experimentally demonstrated at a maximum operating temperature of more than 125 K. Band designs based on two $GaAs/Al_xGa_{1-x}As$ quantum wells with varying aluminum contents of $x = 0.20$, 0.25, and 0.30 were studied. The calculated temperature dependences of the peak gain for two-QW designs with $x > 0.2$ confirm the possibility of creating a THz QCL operating at temperatures above 200 K.

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Authors' contributions

D.V. Ushakov—numerical optimization of THz QCL designs.

A.A. Afonenko—numerical optimization of THz QCL designs.

I.A. Glinskiy—calculation of losses in the THz QCL resonator.

R.A. Khabibullin—fabrication and measurement of the emission characteristics of THz QCLs.

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