# **Quantum cascade laser with bound-to-quasi-continuum optical transitions at a temperature of up to 371 K**

I.S. Molodtsov, N.A. Raspopov, A.V. Lobintsov, A.I. Danilov, A.B. Krysa, I.I. Zasavitskii

Abstract. Based on a matched  $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$  heter**opair, we have developed a quantum cascade laser emitting at a**  wavelength of 7.4  $\mu$ m. The chosen heterostructure with a rela**tively large number of quantum wells and barriers represents two mini-bands separated by a mini-gap with a localised doublet level near the upper mini-band, which provides a wide emission band**   $({\sim}100 \text{ cm}^{-1})$ . In a pulse regime, the maximal laser operation tem**perature is 371 K. Such a high temperature is explained by two factors: a large energy of the transfer from the doublet to the upper mini-band and a large volt defect. The characteristic temperatures**   $T_0$  are found, which are equal to 170 K for low (less than 300 K) **temperatures and 270 K for the range of 300 – 370 K. In addition, optical cavity losses are determined to be 2.5 and 7.7 cm–1 at temperatures of 80 and 254 K, respectively. The pulse power is 0.3 W at 80 K and 0.05 W at 293 K.**

*Keywords: quantum cascade laser, quantum well, potential barrier, superlattice, mini-band, doublet, continuum, MOVPE technique.*

# **1. Introduction**

A quantum cascade laser (QCL) is based on sub-band optical transitions of electrons in a quantum-size heterostructure. The active region structure can be different depending on the desired QCL characteristics [1, 2]. A higher operation temperature can be obtained by increasing the volt defect through the two-phonon (or even three-phonon) electron relaxation from the lower lasing level. However, some practical applications necessitate a higher operation temperature along with a wide spectral range of lasing. From this point of view, the active region is interesting, which has a 'bound-to-continuum state' optical transitions (see [3]). In this case, the upper lasing level is a doublet [4], whereas the lower state is presented by a continuum in which the separation between levels is less than a longitudinal phonon energy. The doublet splitting may be 10–20 meV, which in the case of a doublet –continuum transition broadens the electroluminescence spectrum to 100– 500 cm–1 [5] and gives a chance to obtain a wider QCL gain.

The present work reports about a QCL with doublet – continuum optical transitions, which operates at a wavelength

**I.S. Molodtsov, N.A. Raspopov, I.I. Zasavitskii** Lebedev Physical Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; e-mail: zasavitii@lebedev.ru; **A.V. Lobintsov, A.I. Danilov** Open Joint Stock Company 'M.F. Stel'makh

Polyus Research Institute', ul. Vvedenskogo 3, 117342 Moscow, Russia; **A.B. Krysa** Department of Electronic and Electrical Engineering, University of Sheffield, Mappin Street, Sheffield, S1 3JD, UK

Received 2 March 2020; revision received 27 May 2020 *Kvantovaya Elektronika* **50** (8) 710 –713 (2020) Translated by N.A. Raspopov

of 7.4 um in a wide (up to  $100 \text{ cm}^{-1}$ ) spectral band at temperatures of up to 371 K.

## **2. Laser heterostructure modelling**

The laser structure was constructed by using a  $Ga_{0.47}$  In<sub>0.53</sub>As/  $Al<sub>0.48</sub> In<sub>0.52</sub> As heteropair matched with the lattice constant.$ A single cascade comprised 12 quantum wells (QWs) and the corresponding barriers in the active region: **4.5**/1.6/**1.0**/ 4.7/**1.3**/4.5/**1.3**/4.2/**1.4**/3.5/**1.5**/3.1/**1.6**/3.0/**1.8**/2.8/**2.1**/2.7/**2.5**/ 2.5/**2.9**/2.4/**3.3**/2.2. Here, starting with the injection barrier, the epitaxy layer thicknesses are given in nanometres; bold font refers to barrier thicknesses, conventional font refers to QW thicknesses; doped layers with a concentration of  $1 \times 10^{17}$  cm<sup>-3</sup> are underlined.

Figure 1 presents a diagram of the laser heterostructure calculated for the electric field strength of 62 kV  $cm^{-1}$ . In the diagram, the active region covers the entire cascade and comprises a chirped (with a small intended variation of epitaxy layer thicknesses) superlattice. The chirped structure compensates for an external electric field in the superlattice in order to avoid its doping, which may lead to optical losses. At a sufficiently large number of QW pairs and barriers, the active region is considered as two mini-bands mb1 and mb2 (chirped superlattices) with a mini-gap between those. Chirped superlattice mb1 is a mini-band tilted down which thickness is maximal at the centre and reduces from both sides near injection barriers [3]. In Fig. 1, it is shown as a grey background limiting both superlattices.



**Figure 1.** Calculated energy diagram of a QCL based on a  $Ga_{0.47}In_{0.53}As$  $\overline{Al_{0.48}}$ In<sub>0.52</sub>As heteropair. The intensity of the applied electric field is 62 kV cm–1.

Importantly, at the heteropair parameters chosen, in the mini-gap between mini-bands mb1 and mb2 there is an upper lasing level, which is presented by a doublet D with the splitting of 12 meV. This state is produced by a QW adjacent to the injection barrier. The doublet arises due to anticrossing between the levels of the injector and the QW. Its wavefunction is maximal near the injection barrier and smoothly falls inside the active region. A relatively large doublet splitting provides a wide gain. The splitting can be increased by reducing the injection barrier from 4.5 to 3.7 nm [4, 5].

The upper lasing level is pronouncedly separated (by 75 meV) from high energy states of superlattice mb2, which reduces electron emission to these states and improves the injection efficiency. Electrons tunnel from the ground injector state to the upper doublet level and then are rapidly redistributed between doublet levels due to electron–electron and phonon scattering.

Mini-band mb1 is the lower lasing state. It is a superlattice with a level separation of  $20-25$  meV, which is substantially less than the longitudinal optical phonon energy, and thus, is a kind of a quasi-continuum. Optical transitions occur between the doublet and the quasi-continuum of the lower mini-band (in Fig. 1, these are marked by arrows) with an energy of about 170 meV, which corresponds to the emission wavelength  $\lambda = 7.4$  µm.

The estimated distance from the top of the lower miniband to the injector ground state (volt defect) is above 0.1 meV, which also favours a high temperature of laser operation.

#### **3. Measurement results and discussion**

The laser heterostructures have been grown by the MOVPE method [6] on an *n*-InP substrate. The active region comprised 35 cascades and a relatively large number of QWs and barriers (12 pairs).

A waveguide was produced by surrounding the active region with layers of a three-component  $Ga_{0.47}In_{0.53}As$  solid solution of thickness  $0.23 \mu m$  and a doping level of  $6 \times$ 1016 cm–3, which, in turn, are arranged between two layers of InP with a doping level of  $1 \times 10^{17}$  cm<sup>-3</sup> and thicknesses of  $2 \mu m$  (from the substrate side) and  $2.5 \mu m$  (on top). Figure 2



**Figure 2.** Refractive index profile of the heterostructure and intensity distribution of the fundamental mode in a waveguide.

presents a refractive index profile [7] of the laser heterostructure and intensity distribution for the fundamental waveguide mode. The calculated optical confinement factor is  $\Gamma \approx 0.67$ .

The active element was a mesa-strip construction with a mesa width of  $20 \mu m$ . The crystal was mounted on a copper holder of F-mount type.

For determining cavity losses, lasers with various cavity lengths have been fabricated  $(L = 1-3$  mm) and the threshold current density  $J_{\text{th}}$  has been measured for each of those at temperatures  $T = 80$  and 254 K. The lasers operated in a pulse regime (with the pulse duration of 1 µs and pulse repetition rate of 170 Hz). The integral laser intensity was detected by a photoresistor based on germanium doped with gold or an HgCdTe photodetector. Measurement results are shown in Fig. 3. The measurement error is mainly determined by a small spread of the threshold current density at a prescribed length *L*.



**Figure 3.** Threshold current density  $J_{\text{th}}$  vs. the inverse cavity length at  $T = 80$  and 254 K.

The measured cavity losses were  $2.5 \text{ cm}^{-1}$  at 80 K and 7.7 cm–1 at 254 K. Waveguide losses of radiation in the IR range are conventionally determined by reabsorption, scattering on free carriers, and interface scattering. The obtained waveguide losses substantially increased with temperature, which be can obviously explained by a complicated nature of reabsorption between sub-bands in the employed active region where both vertical and diagonal transitions occur.

The mode gain *gT* determined from Fig. 3 was 7.0 and 6.5 cm kA–1 at temperatures of 80 and 254 K, respectively.

Temperature dependences of the threshold current density were measured for a laser with the cavity length *L =* 3 mm in a close-loop helium refrigerator with a Janis optical cryostat. Measurement results in a wide temperature range  $(40 - 340 \text{ K})$  are presented in Fig. 4. The same dependences measured on another setup at higher temperatures  $(300 - 370 \text{ K})$  are shown in the inset.

The temperature dependence of the threshold current in injection semiconductor lasers is described by the empirical formula  $J_{\text{th}}(T) = J_0 \exp(T/T_0)$ , where  $T_0$  is the characteristic



**Figure 4.** Temperature dependence of the threshold current density  $J_{\text{th}}$ for a OCL  $(L = 3$  mm).

temperature. In our case  $T_0$  corresponds to 170 and 270 K for the low and high temperature ranges, respectively, which is explained by the greater influence of a wide gain at higher temperatures. The change of  $T_0$  temperature in the range from 212 K (at room temperatures) to 510 K (at high temperatures) has been also observed in [5] for lasers on doublet – quasi-continuum transitions at  $\lambda = 8.7 \,\text{\mu m}$ .

The maximal laser operation temperature was 371 K. So a high value was reached due to two factors: a large energy gap between the doublet and upper mini-band mb2, and a large volt defect.

The average integral emission power was measured by using a Vega power metre (Ophir) at the pulse repetition rate increased to 10 kHz. The pulsed power in the multimode operation was 0.3 W at a temperature of 80 K and 0.05 W at 288 K. The results obtained are comparable with those of the first report  $(\lambda = 10 \text{ }\mu\text{m})$  [8] but lower than in later papers on studying QCLs with a similar active region (*l*  $= 8.7 \text{ µm}$ ) [5]. Note that in our case (matched heteropair) the wavelength  $\lambda$  = 7.4 µm corresponds to the transient energy range where the quantum well depth is not large. Hence, in further investigations it is necessary to employ stressed heterostructures.

Emission spectra were recorded by using a Vertex-70 Fourier spectrometer (Bruker) operated in the stepwise scanning regime with a HgCdTe-based photodetector.

Figure 5 shows laser emission spectra near the threshold value  $I_{\text{th}}$  at temperatures of 80 and 288 K. In the first case, the QCL generates at a wavelength of 7.4  $\mu$ m, and in the second case, the emission wavelength is  $7.5 \mu m$ . The measured shift of the emission spectrum maximum is  $46 \text{ cm}^{-1}$  (6 meV), which determines the temperature tuning of the emission wavelength. The mode separation found from spectra of equidistant generation modes is  $\Delta k = 0.51$  cm<sup>-1</sup>. From this follows that the effective refractive index of the active medium is  $N_{\text{eff}}$  = 1/(2 $\Delta kL$ ) = 3.3 at *L* = 3 mm. No noticeable dependence of the effective refractive index on temperature was observed upon recording the emission spectra with a resolution of  $0.2$  cm<sup>-1</sup>.



**Figure 5.** Emission spectra at  $T = 288$  K,  $I = 1.1I_{th}$  and  $T = 80$  K,  $I = 1.2I_{th}$ .

Due to optical transitions between the doublet state and lower mini-band mb1 there is a possibility to obtain a wide gain. At a higher pump current and, respectively, higher Fermi quasi-level, doublet states are gradually filled, which results in a wider emission spectrum of the laser. Thus, the design of the chosen heterostructure allows one to obtain a broadband gain (as compared to the doublet wavelength width). Figure 6 presents emission spectra (of width greater than  $100 \text{ cm}^{-1}$ ) obtained at temperatures of 80 and 288 K. In the first case, the spectrum was recorded by a Fourier spectrometer at a relatively high resolution and high pump current  $(I = 5.5I_{\text{th}})$ ; cavity modes are resolved in the spectrum. The spectrum at  $T = 288$  K was recorded by using a grating monochromator for a laser with a short cavity  $(L = 1 \text{ mm})$ , the pump level was chosen near the threshold value so that the whole wide gain is observed (about  $110 \text{ cm}^{-1}$ ) along with a certain group of modes. So wide a gain is interesting for developing lasers with an external dispersion cavity.



**Figure 6.** Emission spectra at  $T = 80$  K,  $I = 5.5I_{th}$  and  $T = 288$  K,  $I = 1.1I_{\text{th}}$ .

Note that the emission spectrum in Fig. 6 expands to both sides at an increased current. However, in our case the spectrum from the short-wavelength side  $(k > 1340 \text{ cm}^{-1})$  is distorted due to absorption by atmosphere water vapour, because, despite of the falling absorption at longer wavelengths, the path passed by laser emission in atmosphere is above 2 m (both in the monochromator and in the Fourierspectrometer). The emission wavelength of  $7.4 \mu m$  only partially fits the atmosphere absorption band and is of interest in special applications.

## **4. Conclusions**

Based on a  $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$  heteropair, we have developed a quantum cascade laser by using the MOVPE method, which operates at a wavelength of 7.4 µm. The chosen laser heterostructure with a relatively large number of quantum wells and barriers has made it possible to obtain a wide gain (greater than  $100 \text{ cm}^{-1}$ ). The maximal operation temperature in the pulsed regime (371 K) has been obtained due to a high energy of the transition from the doublet level to the upper mini-band and due to a high volt defect value. The characteristic temperature is  $T_0 = 170$  K for temperatures below 300 K and  $T_0 = 270$  K in the range 300–370 K. Optical cavity losses are 2.5 and 7.7 cm–1 at temperatures of 80 and 254 K, respectively. The pulsed emission power is 0.3 W at 80 K and 0.05 W at 288 K. Thus, the developed lasers are interesting for obtaining laser operation at room and higher temperatures in a wide IR spectral range.

*Acknowledgements.* This work was supported by the Presidium of the Russian Academy of Sciences (Actual Problems of Photonics, Probing of Inhomogeneous Media and Materials Programme No. 5.2).

# **References**

- 1. Faist J., Hofstetter D., Beck M., Aellen T., Rochat M., Blaser S. *IEEE J. Quantum Electron*., **38**, 6 (2002).
- 2. Zasavitskii I.I. *Quantum Electron*., **42**, 863 (2012) [*Kvantovaya Elektron*., **42**, 863 (2012)].
- 3. Wittmann A., Gresch T., Gini E., Hvozdara L., Hoyler N., Giovannini M., Faist J. *IEEE J. Quantum Electron*, **44**, 36 (2008).
- 4. Fujita K., Edamura T., Furuta S., Yamanishi M. *Appl. Phys. Lett*., **96**, 241107 (2010).
- 5. Fujita K., Furuta S., Dougakiuch T., Sugiyama A., Edamura T., Yamanishi M. *Opt. Express*, **19**, 2694 (2011).
- 6. Krysa A.B., Roberts J.S., Green R.P., Wilson L.R., Page H., Garcia M., Cockburn J.W. *Cryst. J. Growth*, **272**, 682 (2004).
- 7. Goldberg Yu.A., Schmidt N.M. *Handbook Series on Semiconductor Parameters* (London: World Scientific, 1999) Vol. 2, pp 62–88.
- 8. Maulini R., Beck M., Faist J., Gini E. *Appl. Phys. Lett*., **84**, 1659 (2004).