High performance GaAs/AlGaAs quantum cascade lasers: optimization of electrical and thermal properties

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ABSTRACT

In this paper we present the development of mid-infrared GaAs/AlGaAs QCLs technology and discuss basic characteristics of lasers fabricated at the Institute of Electron Technology. We also show that reliable simulation methods which can deal with the complicated physical phenomena involved in the quantum cascade lasers operation are necessary to predict the behaviour of new structures and optimize their performance. The developed lasers show the record pulse powers of 6 W at 77 K and up to 50 mW at 300 K. This has been achieved by careful optimization of the epitaxial process and by applying a high reflectivity metallic coating to the back facet of the laser. The devices have been successfully used in prototype ammonia detection system working in ppb range.

Keywords: quantum cascade lasers, light-current-voltage characteristics, thermal properties, gas sensing

1. INTRODUCTION

The quantum cascade lasers (QCLs) are unipolar devices based on tunneling and intersubband transitions, in which the electronic states, wavefunctions and lifetimes of relevant states are engineered through the quantum mechanical confinement imposed by a complex multilayer structure. The principle of operation of QCL structures places stringent requirements on the individual layer thickness and composition as well as the overall periodicity of the whole structure. Another crucial problem of QCLs’ operation are the heating effects, which are distinctly larger than in the state-of-the-art bipolar lasers. The heating results in the higher threshold and operation currents of the lasers, and all this in turn results in the necessity of the effective heat extraction. The heat dissipation in QCLs is strongly hampered because of the nature of their active regions containing many interfaces, and layers with thicknesses similar to the mean free path of phonons. The GaAs/AlGaAs quantum cascade lasers were developed shortly after demonstration of first cascade lasers, based on InP1-4. They have been however abandoned due to their inferior performance. Our results show that GaAs/AlGaAs lasers can be developed to the stage of practically useful devices. Bearing in mind that GaAs/AlGaAs technology is by far the most elaborated and cheapest they might still have a commercial potential.

A compact trace-gas detection system for outdoor use, employing developed cascade laser as a light source, has been fabricated5. The mounted laser chip was assembled into a specially designed thermoelectric-cooler box, and its temperature was stabilized to ±0.01K, at 272.2K. By tuning the selected mode of the laser, during the applied current pulse, through an absorption line of the gaseous ammonia around 1046cm⁻¹, a spectroscopic detection of NH₃ was performed with ultrahigh sensitivity. The uncooled MCT detector and 36m multipass optical cell were applied in the system. By application of this cell the ammonia detection with ppb resolution is achievable (~73ppb of ammonia relates to 95% transmission). By use of longer optical path (with commercially achievable multipass cell) even higher resolution is obtainable with the system. The system may be applied also for detection of other types of gaseous substances; e.g., trace amounts of vapors of explosives.

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2. DEVICE DETAILS

The laser structures consist of the 36-period sequence of injector + 3QW segments made of Al$_{0.45}$Ga$_{0.55}$As/GaAs-coupled quantum wells. The active region was based upon the three quantum well design$^6,5$. The layer sequence of one period of the structure, in nanometers, from left to right, starting from the injection barrier is: 4.6, 1.9, 1.1, 5.4, 1.1, 4.8, 2.8, 3.4, 1.7, 3.0, 1.8, 2.8, 2.0, 3.0, 2.6, 3.0 nm. AlGaAs layers are denoted in bold. The total thickness of one period is 45.0 nm. The underlined layers are $n$ doped to preserve charge neutrality of the structure and to prevent formation of high field domains under the applied bias. The optimum injector doping was $\sim 4.4 \times 10^{11}$ cm$^{-2}$. Only two barrier–QW pairs in the central part of each injector have been doped. The structure used a double-plasmon Al-free waveguide for planar optical confinement. The core of the structure was embedded in the lightly doped waveguide composed of 3.5μm thick n-GaAs layers on each side ($n=4.0 \times 10^{16}$ cm$^{-3}$) terminated by 1μm thick highly Si doped ($n=1.0 \times 10^{19}$ cm$^{-3}$) GaAs layers. The laser structures were grown by solid source MBE$^8$.

![Layer sequence in the Al$_{0.45}$Ga$_{0.55}$As/GaAs laser structure](image)

The double trench lasers were fabricated using standard processing technology$^9$, i.e., wet etching and Si$_3$N$_4$ for electrical insulation. The low resistivity Ni/AuGe/Ni/Au ohmic contacts, alloyed in 430°C, were used at the top of the devices. For current injection, windows were opened through the insulator with width 15, 25, and 35 μm. After the wafer was thinned down to about 100 μm, an alloyed AuGe/Ni/Au contact was deposited on the backside. The lasers were cleaved into bars of 1, 1.5, 2 and 3 mm long and soldered with Au/Sn eutectic, epilayer down on diamond heatspreader and copper submounts. An alternative technology, without diamond heatspreader used indium soldering or direct Au/Au bonding. In order to reduce mirror losses, we have developed metallic mirror coatings for high reflectivity. The Al$_2$O$_3$/Au/Al$_2$O$_3$ (100nm/100nm/100nm) HR coatings were applied to the back facet of the laser. Before the coating deposition the laser facet were treated with Ar plasma to remove native oxide. The lasers using this technology show improved performance, i.e., reduced threshold $J_{th}$ and increased slope efficiency.
3. DEVICE MODELING

A very important for the technology development was the ability to predict theoretically the properties of the structures with varied design as well as the possibility to evaluate the influence of a particular design feature on the lasing ability of the devices.

3.1 Electron states in mid-IR GaAs/AlGaAs quantum cascade lasers

The electronic band structure of QCL has been calculated by solving Schroedinger equation with position dependent effective mass by finite difference method (FDM). Injector doping has been taken into account by solving coupled Schroedinger and Poisson equation. The appropriate software package has been developed in Matlab. It allows for calculating relevant properties of the structures depending on their geometry and applied electric field. These include: solving Schroedinger equation for energies and wavefunctions, as well as calculating dipole matrix elements, LO phonon scattering rates, lifetimes and transition rates. The developed software package was primarily used for the structure design, as well as for better understanding of the electrical properties of the achieved QCL structures, and for optimization of device technology.

Fig.2 Conduction band profile and moduli squared wavefunctions in injector/active/injector segment of the laser under the applied field of 48 kV/cm. The E3, E2 and E1 refer to the upper, lower and ground state of lasing transitions.

Fig.3. Conduction band profile and moduli squared wavefunctions in injector/active/injector segment of the laser obtained by solving coupled Schroedinger and Poisson equation for two injector doping levels (2×10^{11} cm^{-2} and 8×10^{11} cm^{-2}).

The essential features of the considered design are diagonal anti-crossed transitions from state E3 to E2 and depopulation of the laser ground state E2 by resonant optical phonon emission and tunneling into the injector (see Fig.2 and Fig.3). The topmost state is the Γ continuum state. It is located ~80 meV above the upper laser level. The calculated lifetime of the excited state and dipole matrix element are τ_{3-1}=1.4 ps, and r_{3}=1.71 nm, respectively. The ground state E2 is depopulated in τ_{2-1} ~ 0.3 ps. These calculations were done at 48 kV/cm, close to estimated laser threshold.

Fig.4. Relative population of electron levels; 4-upper laser level, 2-lower laser level, 1-depopulation level, 3-lowest injector level
Fig. 4 shows results of Monte Carlo calculation of relative population of electron levels involved in laser transitions. The population inversion occurs between level 4 and level 2, but one can notice that the levels in question are occupied only by small fraction of total number of electrons in the system. The majority of electrons resides on the lowest injector level, just before injection barrier.

3.2 Nonequilibrium Green’s function (NEGF) calculations of mid-IR GaAs/AlGaAs quantum cascade lasers

Nonequilibrium Green’s function (NEGF) calculations of mid-infrared quantum cascade laser (QCL) that preserve real-space basis have been performed\(^\text{10}\). The proposed approach relies on two improvements introduced to nonequilibrium Green’s functions/Poisson computational scheme. First, the boundaries of single laser stage were carefully designed as to maintain its periodicity with the whole quantum cascade structure. Second, the NEGF/Poisson solver was equipped with several controlling features that enable the restoration of convergence of the method for quite complex structures with many resonances and boundary conditions for Poisson equation set inside the structure. The model includes band nonparabolicity and takes into account all relevant scattering mechanisms. The model was applied to the QCL emitting at mid-infrared (mIR) frequencies. This structure is quite demanding as the field related to the threshold current is so strong that several (∼7) phonons is emitted by the electron spanning one laser period (see Fig.8). Then, the convergence of NEGF method slows down or even is lost. In the following example, quantities like density of states (Fig.5), electron distribution (Fig.6), current density (Fig.7) and material gain in the structure of QCL were calculated for the case when electrons are scattered by the screened polar-optical phonons (pop) and interface roughness (ir).

![Fig.5. Conduction band profile (solid line) and density of states](image1)

![Fig.6. Conduction band profile and electron distribution in the structure at applied field F = 51 kV/cm](image2)

![Fig.7. Spatio-energetical distribution of current J(z, E) in the laser at applied field F = 51 kV/cm](image3)

![Fig.8. Polar optical (LO\text{Γ}) phonon scattering rate in the laser at applied field F = 51 kV/cm](image4)
The calculated density of states and electron distribution are in quantitative agreement with Monte Carlo results. The energy spectrum of the electrons being injected into upper laser level is concentrated around $E_3$ which means that electrons have small in-plane kinetic energy $E_k = (h k||)^2/2m$. On the other hand the energy spectrum of electrons leaving the active region is multi-picked with spacing equal to $\hbar \omega_{LO} = 36$ meV. This is the result of polar optical phonon scattering of electrons which do not relax by emitting photons. The rate of this process in different sections of the structure is illustrated in Fig.8. The electron distribution in $E_1$ and $E_2$ subbands is strongly nonequilibrium.

NEG F formalism allows for calculating scattering rate for electrons as a function of $z$, $k||$, $E$

$$\Gamma(z, k||, E) = -\frac{2}{\hbar} \text{Im} \left\{ \int_{z'} dz' \exp(i(z - z')\sqrt{2mE / \hbar^2 - k||^2}) \Sigma^R(z, z', k||, E) \right\}$$  \hspace{1cm} (1)

where $\Sigma^R$ is self-energy for given scattering mechanism; in this case polar optical phonon scattering. The electron lifetime for the states involved in laser transitions can be evaluated by averaging with appropriate densities of states.

$$\frac{1}{\tau(E)} = \frac{\int_{z=AR} dz \Gamma(z, 0, E)N(z, 0, E)}{\int_{z=AR} dz N(z, 0, E)}$$  \hspace{1cm} (2)

The function $\tau(E)^{-1}$ is plotted in Fig.9.

![Fig.9. Scattering rate of electrons with $k|| = 0$ in the active region (AR) as a function of total energy.](image)

The lifetime of electrons in the upper laser level and lower laser level are equal $\tau_3 = 1.61$ ps, $\tau_2 = 0.83$ ps, respectively. This is in quite good agreement with $\tau_3 = 1.4$ ps, $\tau_2 = 0.3$ ps, determined by simplified rate equation model discussed in previous section. The differences result from calculation basis; in rate equation model the basis is formed by eigenstates of the system, whereas in this formulation of NEG F method calculation are carried in real-space basis.

NEG F formalism provides a direct way to calculate absorption and gain in the structure. In the first approximation, neglecting variations of scattering rates for particular scattering processes, induced by optical field, gain can be calculated by averaging absorption coefficient over the laser period.

$$g = \frac{1}{\Delta} \int_{z_1}^{z_1+\Delta} dz \alpha(h \nu, z)$$  \hspace{1cm} (3)

The averaging operation allows for determination of the ranges of optical spectrum for which we can expect gain. The laser threshold can be estimated by comparing gain $g$ with total optical losses (waveguide losses plus end losses, divided by confinement factor $\Gamma$). The total optical losses in typical device are of the order of 50 cm\(^{-1}\), while the maximum gain is ~120 cm\(^{-1}\) and exceeds losses in energy range 130-138 meV (see Fig.10a). This is in agreement with experimentally observed emission at 132 meV (9.4 μm). The calculation have been done for 77 K. The width of gain peak results from level broadening and is a consequence of interface roughness scattering. The main contribution to gain broadening comes from upper laser level located in the narrowest well ($d_1 = 1.9$ nm) for which the 1 ML thickness ($\delta = 0.28$ nm) changes $E_3$ level energy much more significantly than it is in the case of the energy levels $E_2$ and $E_1$ located in the thicker wells ($d_2 = 5.4$ nm, $d_3 = 4.8$ nm).
The threshold current density, calculated by integrating $J(z,E)$ over energy $E$, equals $J \approx 4.2 \text{kA/cm}^2$, which is in reasonable agreement with $\approx 5 \text{kA/cm}^2$ determined experimentally. Peak gain vs. current density for different temperatures is shown in Fig.10b. The comparison of calculated electroluminescence (EL) spectrum with measured EL spectrum is shown in Fig.10c.

The results in general correspond to the current understanding of carrier transport in QCL devices; i.e., the electron transport is not purely coherent and one has to take into account scattering as well. NEGF approach is believed to be not only an alternative method for getting numerical solutions of the problems related to various transport phenomena in QCLs, but also a method that provides access and deep insight into the features inaccessible with other, less demanding computationally, simulation tools.

4. EXPERIMENTAL RESULTS

For optimization of electrical and thermal properties of QCLs, several experimental techniques have been used, i.e., temperature dependent Light-Current-Voltage (L-I-V) characteristics, thermoreflectance and spectral characteristics. Lasers with different doping of injectors and different geometries were investigated at varying supply conditions (pulse width and repetition frequency). Of particular interest was maximal operating temperature and power for each device construction.

4.1 Light-Current-Voltage (L-I-V) characteristics

The current-voltage characteristics, obtained for the structures in which the thickness and composition of active region layers were correctly engineered, are characterized by specific features (see Fig.11a). At low bias QCL structures should be highly resistive. When the electric field reaches the value for which the alignment of the upper laser state and the injector ground state takes place, the resistance of the structure drops down and electrons start flowing through the device. In this regime the operating voltage increases linearly with injection current. The saturation of the V-I
characteristics, i.e., the high differential resistance of the device, is caused by the onset of misalignment between the upper laser level and the injector ground state. The above description should match the results of V-I measurements performed at low temperature (77K); for higher temperatures the lower values of device differential resistance at all current regimes are observed are as a rule.

The effect of saturation, limiting the dynamic range of laser operation depends on the injector doping concentration. As the threshold condition may be reached only after the gain exceeds the losses, it is possible that the saturation condition may appear before the losses are exceeded. That is why to achieve lasing the high enough currents must flow through the structure before the saturation, and this requirement has to be fulfilled by a proper injector doping. A range of injector doping was tested for a fixed QCL geometry. It turned out that optimum injector doping was 2x10^{11} cm^{-2} - 4.4x10^{11} cm^{-2}. For the electro-optical measurements QCLs were placed in liquid nitrogen cryostat, allowing for temperature changes from 77K up to room temperature (300 K). Devices were driven with 200 ns pulses of 1 kHz frequency for standard tests. The other driving conditions (pulse frequency up to 5 kHz and pulse length up to 10 µs) were used for specific tests.

The characteristic feature of QCLs is optical power rollover at high currents^{11,12}. Contrary to what is observed in conventional diode laser this is a completely reversible phenomenon, having nothing in common with device degradation. The rollover observed in (L-I) curves of narrow ridge lasers (see Fig.11a) can be explained in terms of two effects, the thermal rollover (the positive feedback loop between increasing laser core temperature and threshold current density) and Stark shift of the laser levels causing their misalignment with increasing current (Stark rollover). For low temperatures of heat sink (77 K-140 K) a voltage step in the current-voltage curve, corresponding to a rapid drop in optical power, is observed. It occurs at supply current 3.4 A (current density 15kA/cm^2) and voltage ~ 13V. The current at which a maximum available power is attained doesn’t change which is characteristic for Stark rollover. While increasing temperature (190K-270K), the transition from Stark rollover to thermal rollover is observed. The characteristic feature of thermal rollover is that the drop of peak power occurs at progressively increasing current and is less abrupt than in the previous case. The devices with wider ridges (see Fig.11b) exhibit pure thermal rollover because they need higher current to lase and consequently the start overheating before they get misaligned. In that regime the current increase results in much smaller voltage (electric field) change over the laser core as evidenced by high temperature (I-V) characteristics. The output power scales linearly, at least to certain dimensions, with the volume of laser active region. This is demonstrated in Fig.12 which shows light–current and current-voltage characteristics of the GaAs/Al_{0.45}Ga_{0.55}As laser driven by 200 ns pulses with repetition rate of 5 kHz. The laser cavity length equals 2 mm and the mesa width equals 35 μm. The output power of 5 W at 77 K is achieved and the device does not show signs of rollover. The wall plug efficiency reaches 7% and the external differential efficiency is ~ 1 W/A (8 photons are emitted by 1 electron on average).
The parameters of developed lasers are listed in Table 1. The total number of a few hundreds devices have been fabricated and tested; among them ~ 200 fully assembled. The parameters given in Table 1 refer to devices with optimized injector doping and optimized processing (III-processing session).

### Table 1. GaAs/AlGaAs quantum cascade laser parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Threshold current density</td>
<td>$J_{th}(77K)$</td>
<td>$5 \text{kA/cm}^2 - 6 \text{kA/cm}^2$</td>
</tr>
<tr>
<td>Threshold current density</td>
<td>$J_{th}(300K)$</td>
<td>$10 \text{kA/cm}^2 - 15 \text{kA/cm}^2$</td>
</tr>
<tr>
<td>Threshold voltage</td>
<td>$V_{th}(77K)$</td>
<td>~ 8.5 V</td>
</tr>
<tr>
<td>Power</td>
<td>$P(77K)$</td>
<td>&gt; 4 W (max 6 W)</td>
</tr>
<tr>
<td>Power</td>
<td>$P(300K)$</td>
<td>25 mW – 35 mW</td>
</tr>
<tr>
<td>Emission wavelength</td>
<td>$\lambda(300K)$</td>
<td>~ 9.4 $\mu$m - 9.5 $\mu$m</td>
</tr>
<tr>
<td>Characteristic temperature</td>
<td>$T_0$</td>
<td>~ 100 K</td>
</tr>
<tr>
<td>Series resistance</td>
<td>$R_s(77K-300K)$</td>
<td>0.5 $\Omega$ – 0.7 $\Omega$</td>
</tr>
<tr>
<td>External differential efficiency</td>
<td>$\eta_d(77K)$</td>
<td>0.8 W/A - 1.2 W/A</td>
</tr>
<tr>
<td>Wall plug efficiency</td>
<td>$\eta_{wpe}(77K)$</td>
<td>6 % - 8 %</td>
</tr>
</tbody>
</table>

### 4.1 Thermal characterization

In order to gain insight into the thermal properties of QCLs the thermoreflectance (TR) spectroscopy was used. TR is an optical modulation technique, which relies on measurement of the relative change in reflectivity induced by periodic change of the sample’s temperature\cite{13-15}. A probe laser beam incidents perpendicularly to the facet and is reflected back. The periodic temperature change of the laser induces variations of the refractive index and consequently modulates the probe beam reflectivity. The TR mapping has been successfully applied to study heating effects in quantum cascade lasers\cite{16}.

![Fig.12. Light–current and current-voltage characteristics of the GaAs/Al$_{0.45}$Ga$_{0.55}$As laser driven by 200 ns pulses with repetition rate of 5 kHz. The laser cavity length equals 2 mm and the mesa width equals 35 $\mu$m.](image-url)
Fig. 13 presents high resolution maps of the temperature distribution recorded over 90\(\mu\)m x 100\(\mu\)m area for different supply conditions; i.e., current, pulse frequency and duration. As, contrary to interband lasers, emitted radiation is not absorbed at the mirror surface, that temperatures represent true temperature inside the device.

The main heat source is the active region; no heat generation has been observed at the contact. In general, the epi-down mounted lasers do not overheat. The highest temperature increase, observed for long 50 \(\mu\)s pulses was \(\Delta T = 30\)K. For typical supply conditions (200 ns, 5 kHz) no difference in thermal behavior of epi-up and epi-down mounted lasers was observed. The devices' thermal resistances are derived from the experimental data. Their typical values are \(\sim 20\) K/W.

We have also developed numerical thermal model of QC lasers solving heat transport equation in 2D and 3D, which includes anisotropy of thermal conductivity and the influence of doping. By combining the experimental and numerical results, an insight into the thermal management in QCLs has been gained. The thermal optimization of the design focused on improving heat dissipation in the device, which is essential to increase the maximal operation temperature of the devices.

5. CONCLUSIONS

The development of (\(\lambda\sim 9.4\mu\)m) GaAs-based quantum cascade lasers (QCLs) operating up to 300 K is reported. This has been achieved by the use of GaAs/Al\(_{0.45}\)Ga\(_{0.55}\)As heterostructures. The laser design followed an 3QW anticrossed-diagonal scheme. The QCL structures were grown by MBE, with Riber Compact 21T reactor. The peak powers recorded in 77K were over 4 W (25 mW – 35 mW at 300 K), and the slope efficiency \(\eta = 0.5-0.6\) W/A per uncoated facet (~1 W/A
for lasers with metallic back mirror coating). These results are comparable with the state of the art GaAs/AlGaAs devices of similar design produced in other laboratories\textsuperscript{17,18}. The lasers have been successfully used in prototype ammonia detection system working in ppb range. Our results show that GaAs/AlGaAs lasers can be developed to the stage of practically useful devices. Bearing in mind that GaAs/AlGaAs technology is by far the most elaborated and cheapest they might still have a commercial potential.

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