Quantum Cascade Lasers for the Mid-infrared Spectral Range: Devices and Applications

Ch. Mann¹, Q. K. Yang¹, F. Fuchs¹, W. Bronner¹, R. Kiefer¹, K. Köhler¹, H. Schneider¹, R. Kormann², H. Fischer², T. Gensty³, and W. Elsäßer³

¹ Fraunhofer-Institute for Applied Solid State Physics (IAF)
Tullastrasse 72, D-79108 Freiburg, Germany
christian.mann@iaf.fraunhofer.de

² Max-Planck-Institute for Chemistry
J.J. Becher-Weg 27, D-55128 Mainz, Germany

³ Institute of Applied Physics, Darmstadt University of Technology
Schlossgartenstrasse 7, D-64289 Darmstadt, Germany

Abstract. Quantum cascade lasers emitting at \( \lambda \sim 5 \mu m \) based on different active region designs are investigated. Using lattice-matched GaInAs/AlInAs on InP substrates the maximum peak optical power as well as the maximum pulsed-mode operating temperature is enhanced by incorporating AlAs blocking barriers together with strain-compensating InAs layers into the active regions. Further improvement is achieved by employing strain-compensated GaInAs/AlInAs quantum wells for which maximum pulsed-mode operating temperatures in excess of 350 K are observed. High-reflectivity coated devices mounted substrate-side down show a maximum continuous-wave operating temperature of 194 K. Also the normalized relative intensity noise is investigated. Finally, a comparison trace-gas sensing experiment employing one of the present quantum cascade lasers and a lead-chalcogenide laser is presented. Detecting the P(25) absorption line of CO, higher stability is obtained using a quantum cascade laser.

1 Introduction

Quantum cascade (QC) lasers are unipolar mid- to far-infrared emitters in which the laser transition occurs between quantized energy levels within e.g., the conduction band. As the emission wavelength is determined by quantum confinement, a broad wavelength range can be covered by tuning the thicknesses of the individual layers without changing the material compositions. Since their first demonstration in 1994 employing GaInAs/AlInAs grown on InP substrate [1] these novel devices have made tremendous progress. At present the wavelength range covered by this material system reaches from 3.5 \( \mu m \) [2] to 24 \( \mu m \) [3] and also two-wavelength [4,5] as well as broadband QC lasers emitting continuously between 6 \( \mu m \) and 8 \( \mu m \) were fabricated [6]. A maximum pulsed-mode operating temperature of 470 K at a wavelength of 5.5 \( \mu m \) [7] and recently continuous-wave (CW) operation at room temperature for devices emitting at 9.1 \( \mu m \) [8] was reported. Because of the high
operating temperature and the high pulsed optical power QC lasers are getting suitable light sources for trace-gas sensing as well as for optical free-space communication. However, for devices emitting at around 5 µm wavelength the maximum CW operating temperature is currently still limited to 210K [9].

To achieve population inversion necessary for laser action the lifetime of electrons in the final state of the laser transition must be shorter than the lifetime of electrons in the initial state. For this reason, in QC lasers resonant emission of longitudinal-optical (LO) phonons from the final state to a bound state lying approximately one LO-phonon energy below is widely employed as depopulation mechanism [1]. Under operating conditions electrons are injected into the active region of the QC laser structure by resonant tunneling through the injection barrier. After making radiative transitions they relax by resonant LO-phonon emission and tunnel through the exit barrier into the following injector. The injectors are designed such that a minigap opens up for electrons in the initial state of the laser transition to prevent them from leaking into the injector (“Bragg reflector”), while a miniband is created for electrons in the final states favoring them to tunnel through to be injected into the active region of the next period [10,11,12].

In the present paper we focus on GaInAs/AlInAs/InP based QC lasers emitting in the 5 µm wavelength range. First, modified designs aiming at enhancing population inversion are investigated. In Sect. 2 we show that the confinement of electrons in the initial state of the laser transition of QC lasers using lattice-matched GaInAs/AlInAs is enhanced by incorporating AlAs blocking barriers together with strain-compensating InAs layers into the active regions. In Sect. 3 we report on QC lasers employing strain-compensated active regions. For these devices the increased conduction band offset further enhances the electrical confinement and additionally offers more design flexibility. Next, in Sect. 4 the normalized relative intensity noise is investigated. In Sect. 5 a comparison trace-gas sensing experiment employing one of the present QC lasers and a lead-chalcogenide laser detecting the P(25) absorption line of CO is presented and finally, in Sect. 6 a brief summary is given.

2 Advantage of Blocking Barriers in the Active Regions of λ ~ 5 µm Quantum Cascade Lasers

In this section we report on the improvement of λ ~ 5 µm QC lasers by incorporating AlAs blocking barriers together with strain-compensating InAs layers into the active regions [13]. With respect to a reference sample without blocking barriers the maximum peak optical power at 77 K is increased by a factor of 3. Additionally, the maximum pulsed-mode operating temperature is increased from 320 K to 350 K.
2.1 Concept of Blocking Barriers

The calculated conduction band profile (Γ-valley) of two active regions connected by an injector of a QC laser structure designed for emission at $\lambda \sim 5 \mu m$ using Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As grown lattice-matched on InP substrate, as published in [12], is shown in Fig. 1a. In this design the injector acts as a “Bragg reflector” for electrons in the initial state of the laser transition (level 3), whereas electrons in the final states (level 2 and level 1) are favored to tunnel through the exit barrier into the injector’s miniband as described in Sect. 1. The detailed layer thicknesses and doping profiles are described in the caption of Fig. 1.

To further enhance the confinement of electrons in level 3, the 3.0nm Al$_{0.48}$In$_{0.52}$As exit barrier was substituted by a triple layer sequence composed of 0.7nm AlAs (“blocking barrier”) sandwiched between 0.9nm Al$_{0.48}$In$_{0.52}$As (see Fig. 1b) [13]. The tunneling probability of electrons in a state $|i>$ of a quantum well through a barrier of thickness $L_b$ is proportional to $\exp(-2L_b\sqrt{2m_bV_i/\hbar})$ with the effective mass of electrons in the barrier $m_b$, the effective barrier height $V_i$ and Planck’s constant $\hbar$. For this reason, the direct tunneling/emission of electrons in level 3 into the next active region/continuum is selectively blocked by increasing $V_3$. At the same

![Fig. 1. Schematic conduction band profile (Γ-valley) of two active regions connected by an injector under positive bias condition at an electric field of 75 kV/cm. Also shown are the moduli squared of the relevant wave functions and the first miniband of the injector (grey shaded region). The laser transition is indicated by wavy arrows. (a) Structure as published in [12]. The layer sequence of one active region and injector, in nanometers, from left to right starting from the injection barrier is 5.0, 0.9, 1.5, 4.7, 2.2, 4.0, 3.0, 2.3, 2.2, 2.2, 2.0, 2.0, 2.0, 2.3, 1.9, 2.8, 1.9. (b) Structure with incorporated AlAs blocking barriers. The layer sequence of one active region and injector, in nanometers, from left to right starting from the injection barrier is 5.0, 1.0, 1.5, 2.0, 0.7 (InAs), 2.0, 2.2, 4.1, 0.9, 0.7 (AlAs), 0.9, 2.5, 2.3, 2.3, 2.2, 2.0, 2.0, 2.0, 2.3, 1.9, 2.8, 1.9. The Al$_{0.48}$In$_{0.52}$As barriers (Ga$_{0.47}$In$_{0.53}$As quantum wells) are typeset in bold (roman), layers in italic are Si doped to $n = 2 \times 10^{17}$ cm$^{-3}$, and the underlined layers serve as the exit barrier.
time the high tunneling probability out of level 2 and level 1 is maintained by reducing the total width of the triple layer exit barrier, such that the products $L_b \sqrt{2m_b V_2}$ and $L_b \sqrt{2m_b V_1}$ are kept essentially unchanged.

For the QC laser structure shown in Fig. 1a level 3 is close to the conduction band edge of the Al$_{0.48}$In$_{0.52}$As exit barrier and consequently the effective barrier height $V_3$ is small. By incorporating AlAs blocking barriers into the active regions the calculated tunneling probability out of level 3 is reduced by a factor of two ($2L_b \sqrt{2m_b V_3}/\hbar = 1.8$ for the design of Fig. 1b and $2L_b \sqrt{2m_b V_3}/\hbar = 1.1$ for the design of Fig. 1a). As the total width of the exit barrier is reduced from 3.0 nm to 2.5 nm the high tunneling probability out of level 2 and level 1 is maintained ($2L_b \sqrt{2m_b V_2}/\hbar = 2.9$ and $2L_b \sqrt{2m_b V_1}/\hbar = 3.0$ for both designs).

To compensate for the strain caused by introducing AlAs blocking barriers thin InAs layers were additionally incorporated into the active quantum well. Furthermore, these layers increase the depth of the effective potential, thus allowing more design flexibility towards shorter emission wavelengths.

From now on wafers based on the design of Fig. 1a (Fig. 1b) will be referred to as sample A (sample B). At an applied electric field of 75 kV/cm the calculated transition energy of sample A (sample B) is 250 meV (258 meV), corresponding to an emission wavelength of 5.0 $\mu$m (4.8 $\mu$m). For both designs the energy difference between level 2 and level 1 is approximately equal to one LO-phonon energy for the purpose of LO-phonon assisted depopulation of the final state of the laser transition (see Sect. 1). The calculated LO-phonon scattering times of sample A (sample B) are $\tau_{32} = 2.3$ ps (3.7 ps), $\tau_{31} = 2.8$ ps (2.5 ps) yielding a lifetime $\tau_3 = 1.3$ ps (1.5 ps) with $\tau_3^{-1} = \tau_{32}^{-1} + \tau_{31}^{-1}$ of the initial state, and $\tau_{21} = 0.3$ ps (0.2 ps). The transition matrix-element is $|z_{32}| = 1.7$ nm (1.4 nm).

## 2.2 Device Fabrication

For both designs shown in Fig. 1 twenty-five periods of alternating active regions and injectors embedded between 400 nm thick Si-doped ($n = 1 \times 10^{17}$ cm$^{-3}$) Ga$_{0.47}$In$_{0.53}$As separate confinement layers were grown lattice-matched on S-doped ($n = 2 \times 10^{17}$ cm$^{-3}$) (001)-oriented InP substrates by molecular beam epitaxy (MBE). Then the wafers were transferred to a metal-organic chemical vapor deposition (MOCVD) system, where Si-doped InP serving as upper waveguide and contact layers was grown (20 nm, $n = 5 \times 10^{17}$ cm$^{-3}$; 1500 nm, $n = 2 \times 10^{17}$ cm$^{-3}$; 1300 nm, $n = 7 \times 10^{18}$ cm$^{-3}$). For both samples we calculate values of $n_{eff} = 3.23$ for the effective mode refractive index, $\Gamma_{tot} = 0.55$ for the mode confinement factor considering the mode overlap with active regions and injectors, and $\Gamma = 0.26$ for the mode confinement factor considering the mode overlap with active regions only.

The wafers were processed into 8–16 $\mu$m wide ridge-waveguide structures by chemical assisted ion beam etching (CAIBE) to a depth of 4 $\mu$m. Then
a 350 nm Si$_3$N$_4$ passivation layer was deposited by plasma enhanced chemical vapor deposition (PECVD), windows were opened on top of the ridges, and Ge/Ni/Ge/Ni/Au (5/5/5/5/400 nm) was evaporated as the top contact metallization. After thinning the substrates to approximately 110 µm thickness the same metallization was deposited as the backside contact. Finally, lasers of 1–3 mm length were cleaved from the processed wafers and mounted substrate-side down with uncoated facets on copper heat-sinks.

For all measurements presented in Sects. 2 and 3 the samples were placed inside a temperature controlled continuous flow cryostat. The optical power emitted into a solid angle of about $\pi/10$ was detected with a calibrated liquid nitrogen cooled InSb detector using calibrated attenuation filters to avoid saturation. Emission spectra were analyzed with a Bomem DA3 Fourier transform spectrometer.

### 2.3 Device Performance

Prior to comparing the device performance of QC lasers with and without AlAs blocking barriers the modal gain of reference sample A without blocking barriers is investigated.

Employing the method proposed by Hakki and Paoli, the net modal gain defined as $\Gamma g - \alpha_i$, with the mode confinement factor $\Gamma$, the material gain $g$, and the internal losses $\alpha_i$, can be determined experimentally from sub-threshold electroluminescence (EL) spectra [14,15]. Figure 2a shows net modal gain spectra of an $8 \times 1000 \mu$m$^2$ device of sample A at a heat-sink temperature of 30 K for various injection currents between 120 mA and 210 mA. The net modal gain maximum occurs at an emission energy of about 1955 cm$^{-1}$ (5.1 µm). The peak net modal gain as a function of injection

![Fig. 2.](attachment:fig2.png)

(a) Measured net modal gain of an $8 \times 1000 \mu$m device of sample A at a heat-sink temperature of 30 K for various injection currents between 120 mA and 210 mA. (b) Peak net modal gain as a function of current density (squares) and linear least square fit to the experimental data (solid line). The threshold condition $\Gamma g_{\text{peak}}(J_{\text{th}}) - \alpha_i = \alpha_m$ is indicated (circle)
current density, plotted in Fig. 2b, shows a linear dependence with a slope \( \Gamma g_0 = (9.1 \pm 0.2) \text{ cm/kA} \). As can be seen from extrapolating the experimental data to the threshold condition \( \Gamma g_{\text{peak}}(J_{\text{th}}) - \alpha_i = \alpha_m \), with the measured threshold current density \( J_{\text{th}} = 2.85 \text{kA/cm}^2 \) and the calculated mirror loss \( \alpha_m = 13.1 \text{ cm}^{-1} \), no gain saturation occurs. Extrapolation to \( J = 0 \) yields internal losses of \( \alpha_i = (12.4 \pm 0.5) \text{ cm}^{-1} \).

Theoretically, gain spectra can be calculated from Fermi’s golden rule [16]. Assuming perfectly parabolic subbands and a Lorentzian line shape, the peak material gain \( g_{\text{peak}}(J) = g_0 J \) is proportional to the injection current density \( J \), and the gain coefficient \( g_0 \) reads for unity injection efficiency into the initial state of the laser transition [11,17]

\[
g_0 = \frac{4\pi e |z_{32}|^2}{\varepsilon_0 n_{\text{eff}} \lambda_0 L_p (2\gamma_{32})} \tau_3 \left(1 - \frac{\tau_{21}}{\tau_{32}} \right),
\]

In (1) \( e \) is the electron charge, \( \varepsilon_0 \) is the dielectric constant, \( n_{\text{eff}} \) is the effective mode refractive index, \( \lambda_0 \) is the free-space wavelength corresponding to the gain maximum, \( L_p \) is the length of one active region/injector stage and \( (2\gamma_{32}) \) is the full width at half maximum of the EL. Using (1) together with \( (2\gamma_{32}) = 15 \text{ meV} \) and the parameters of Sects. 2.1 and 2.2, a value of \( \Gamma g_0 = 10.5 \text{ cm/kA} \) is obtained in reasonable agreement with experiment.

The light output versus injection current (L-I) dependence at various heat-sink temperatures and the voltage versus injection current (V-I) characteristic at 300 K is shown in Fig. 3a (Fig. 3b) for a 16 \times 3000 \mu m^2 device of sample A (sample B). The lasers are driven by current pulses of 100 ns length at a repetition rate of 5 kHz. For both samples the operating voltage is approximately 7 V. The peak optical power of reference sample A at 77 K increases almost linearly with injection current above threshold \( I_{\text{th}} = 0.9 \text{ A} \) up to \( I = 2.5 \text{ A} \) and \( P = 250 \text{ mW} \). The maximum optical power of 285 mW is reached at an injection current of 3.6 A, for higher injection current a rollover of the optical power appears. A similar behavior of the (L-I)-dependence is found at elevated heat-sink temperatures. At 300 K the maximum optical power is 30 mW at \( I = 3.6 \text{ A} \). The maximum pulsed-mode operating temperature is 320 K. In comparison, the device of sample B with incorporated blocking barriers operates at higher injection current and thus at higher optical power. At 77 K (300 K) the (L-I)-dependence is almost linear from threshold \( I_{\text{th}} = 0.8 \text{ A} (I_{\text{th}} = 3.4 \text{ A}) \) up to an injection current of 5.0 A (5.5 A) with a maximum optical power of 890 mW (245 mW). For higher injection current the optical power decreases rapidly. The maximum operating temperature is 350 K.

There are two mechanisms responsible for the rollover of the optical power with increasing injection current. (1) Since more electrons are injected into level 3, at a certain injection current the confinement of these electrons becomes insufficient and they start leaking out of the active region, resulting in a plateau in the (L-I)-curve. (2) Due to the increasing applied voltage the energy levels in the active regions and injectors become misaligned and the
Fig. 3. Light output versus injection current (L-I) dependence at various heat-sink temperatures (solid) and voltage versus injection current (V-I) characteristic at 300 K (dashed) of (a) sample A and (b) sample B. The $16 \times 3000 \mu m^2$ devices are driven by current pulses of 100 ns length at a repetition rate of 5 kHz.

Resonant tunneling/injection is interrupted, giving rise to a sharp decrease of the optical power. For reference sample A mechanism (1) gives rise to the observed plateau in the (L-I)-curve. We attribute the ability of sample B to operate at higher injection current, and thus at higher optical power (about a factor of 3 at 77 K), to the fact that mechanism (2) sets in at higher injection current than mechanism (1), due to the significantly enhanced confinement of electrons in the initial state of the laser transition by incorporating blocking barriers into the active regions.

For both sample A and sample B the threshold current increases exponentially with a characteristic temperature $T_0 = 136$ K. At elevated temperatures the threshold current of sample B is about 20% higher compared to sample A, which might be caused by a slightly broadened gain spectrum and a reduced transition matrix-element (see Sect. 2.1). The peak emission wavelength of sample A (sample B) at 77 K is 5.05 $\mu$m (4.94 $\mu$m) [13]. The blueshift of the emission of sample B is attributed to the increased depth of the effective potential due to the incorporated strain-compensating InAs layers (see Sect. 2.1).
3 Quantum Cascade Lasers Based on Strain-Compensated GaInAs/AlInAs

In this section we show that the device performance can be further improved by using strain-compensated GaInAs/AlInAs in the active regions and injectors, giving rise to an increased conduction band offset. Employing a modified design based on “double-phonon relaxation” [18] maximum pulsed-mode operating temperatures in excess of 350 K are observed. High-reflectivity coated samples mounted substrate-side down show a maximum CW operating temperature of 194 K.

3.1 Sample Design

To further enhance the electrical confinement of electrons in the active regions and to achieve more design flexibility towards shorter emission wavelengths, the conduction band offset can be increased by using strained Ga$_{1-x}$In$_x$As quantum wells ($x > 0.53$) and Al$_{1-y}$In$_y$As barriers ($y < 0.52$). As the individual layers in the active regions and injectors of QC lasers are sufficiently thin (typically below 5 nm) they remain below the critical layer thickness. The compositions and layer thicknesses are chosen such that the compressive strain introduced by the quantum wells is compensated by the tensile strain introduced by the barriers. Using Al$_{0.6}$In$_{0.4}$As/Ga$_{0.38}$In$_{0.62}$As the conduction band offset is increased to $\sim 710$ meV compared to $\sim 510$ meV for lattice-matched Al$_{0.48}$In$_{0.52}$As/Ga$_{0.47}$In$_{0.53}$As.

Figure 4 shows the calculated conduction band profile (Γ-valley) of two active regions connected by an injector of a QC laser structure based on strain-compensated Al$_{0.6}$In$_{0.4}$As/Ga$_{0.38}$In$_{0.62}$As. The design employs a four quantum well active region with three lower bound states (levels 1–3) separated by one LO-phonon energy each [18]. Due to the “double-phonon relaxation” the extraction efficiency out of the active region into the following injector is enhanced, thus reducing the lifetime of the final state of the laser transition (level 3), and thermal backfilling of electrons from the injector into the final state is reduced, thus improving the high-temperature performance. Additionally, the confinement of electrons in the initial state (level 4) is enhanced by the increased potential depth. Also AlAs blocking barriers together with strain-compensating InAs layers (see Sect. 2) are incorporated into the active regions. The detailed layer thicknesses and doping profiles are described in the caption of Fig. 4. At an applied electrical field of 75 kV/cm the calculated transition energy is 272 meV, corresponding to an emission wavelength of 4.6 μm. The estimated LO-phonon scattering time of level 4 (level 3) is $\tau_4 = 1.1$ ps ($\tau_3 = 0.2$ ps) using $\tau_4^{-1} = \tau_4^{-1} + \tau_4^{-1} + \tau_4^{-1}$ and $\tau_3^{-1} = \tau_3^{-1} + \tau_3^{-1}$ with $\tau_4 = 2.8$ ps, $\tau_3 = 2.9$ ps, $\tau_4 = 5.6$ ps, $\tau_3 = 0.3$ ps and $\tau_3 = 1.7$ ps. For the transition matrix-element a value of $|z_{43}| = 1.8$ nm is determined.
Fig. 4. Schematic conduction band profile (Γ-valley) of two active regions connected by an injector under positive bias condition at an electric field of 75 kV/cm. Also shown are the moduli squared of the relevant wave functions and the first miniband of the injector (grey shaded region). The laser transition is indicated by wavy arrows. The layer sequence of one active region and injector, in nanometers, from left to right starting from the injection barrier is 4.4, 0.9, 1.1, 4.8, 1.5, 4.4, 1.6, 1.6, 0.6 (InAs), 1.6, 0.9, 0.6 (AlAs), 0.9, 3.1, 1.2, 2.9, 1.4, 2.8, 1.6, 2.7, 2.0, 2.5, 2.3, 2.3, 2.1, 3.0.1.9. The Al$_{0.6}$In$_{0.4}$As barriers (Ga$_{0.38}$In$_{0.62}$As quantum wells) are typeset in bold (roman), layers in italic are Si doped to $n = 3 \times 10^{17}$ cm$^{-3}$, and the underlined layers serve as the exit barrier.

Samples based on the design of Fig. 4 were grown and processed as described in Sect. 2.2 with the following exceptions. The silicon doping profiles of the upper Ga$_{0.47}$In$_{0.53}$As separate confinement layer (300 nm, $n = 1 \times 10^{17}$ cm$^{-3}$; 200 nm, $n = 2 \times 10^{17}$ cm$^{-3}$) as well as of the MOCVD grown InP upper waveguide and contact layers (20 nm, $n = 5 \times 10^{17}$ cm$^{-3}$; 1500 nm, $n = 2 \times 10^{17}$ cm$^{-3}$; 1200 nm, $n = 2 \times 10^{18}$ cm$^{-3}$; 100 nm, $n = 7 \times 10^{18}$ cm$^{-3}$) were modified. The etch depth was 4.7 µm, and Ge/Ni/Ge/Ni/Au (5/5/5/5/300 nm) followed by Ti/Au (50/400 nm) was evaporated as the top contact.

3.2 Device Performance

The light output versus injection current (L-I) dependence at various heatsink temperatures and the voltage versus injection current (V-I) characteristic at 300 K is shown in Fig. 5a for a 16×3000 µm$^2$ device driven by current pulses of 100 ns length at a repetition rate of 5 kHz. The operating voltage is slightly above 9 V. The device shows further improved performance compared to the QC lasers presented in Sect. 2. At 240 K the maximum peak power (slope efficiency) is 846 mW (655 mW/A) decreasing to 473 mW (400 mW/A) at 300 K and to 97 mW (132 mW/A) at 350 K, which is the maximum temperature achievable with our setup. Therefore higher operating temperatures are possible. The threshold current density $J_{th}$ as a function of heat-sink temperature $T$ increases exponentially from 1.9 kA/cm$^2$ at 240 K to 3.1 kA/cm$^2$ at 300 K.
Fig. 5. (a) Light output versus injection current (L-I) dependence at various heat-sink temperatures (solid) and voltage versus injection current (V-I) characteristic at 300 K (dashed). The 16 × 3000 µm² device is driven by current pulses of 100 ns length at a repetition rate of 5 kHz. (b) Normalized pulsed-mode emission spectra at various heat-sink temperatures and to 4.5 kA/cm² at 350 K. Fitting the experimental data to the empirical relation $J_{th}(T) = J_0 \exp(T/T_0)$, a value of $J_0 = 0.31$ kA/cm² and a characteristic temperature $T_0 = 131$ K is determined. Normalized pulsed-mode emission spectra at various heat-sink temperatures are displayed in Fig. 5b.

As expected for Fabry-Perot devices in pulsed-mode operation, the emission spectra are multi-mode with a longitudinal mode spacing of $\sim 0.5 \text{ cm}^{-1}$. The peak of the emission shifts from 2049 cm$^{-1}$ (4.88 µm) at 240 K to 2024 cm$^{-1}$ (4.94 µm) at 350 K, in reasonable agreement with the calculated transition energy.

In order to reduce the threshold current density and the injected electrical power leading to improved CW performance, the devices can be high-reflectivity (HR) coated. As HR-coated lasers based on the design shown in Fig. 4 are not yet available, we present our results on HR-coated lasers based on similar design, but employing a triple quantum well active region with single-phonon relaxation as published in [19] next. For the back facet two $\lambda/4$ pairs of SiO$_2$/Si resulting in a reflectivity of $\sim 96\%$ were deposited. The front facet was coated with one $\lambda/4$ pair of SiN/Si giving rise to a reflectivity of $\sim 65\%$. After coating the devices were mounted substrate-side down on copper heat-sinks.

Figure 6a shows the CW light output versus injection current (L-I) dependence at various heat-sink temperatures and the voltage versus injection current (V-I) characteristic at 82 K of an 8 × 1000 µm² device. The operating voltage is slightly above 8 V. At a heat-sink temperature of 82 K the threshold current is 46 mA and a maximum optical power of almost 80 mW is emitted. At the maximum CW operating temperature of 194 K the threshold current is increased to 279 mA with the maximum optical power still exceeding 1 mW. We attribute the noise in some parts of the L-I-curves to spatial mode hop-
Quantum Cascade Lasers for the Mid-infrared Spectral Range

Fig. 6. (a) CW light output versus injection current (L-I) dependence at various heat-sink temperatures (solid) and voltage versus injection current (V-I) characteristic at 82 K (dashed) of an $8 \times 1000 \mu m^2$ device with HR-coated facets mounted substrate-side down. (b) Normalized CW emission spectra ($I \approx 1.1 I_{th}$) at various heat-sink temperatures

...ping of the Fabry-Perot device. The maximum CW operating temperature is close to the highest published value of 210 K for QC lasers at $\lambda \sim 5 \mu m$ obtained for a junction down mounted $12 \times 2000 \mu m^2$ device with uncoated facets [9]. Normalized CW emission spectra at various heat-sink temperatures are displayed in Fig. 6b for injection currents $I \approx 1.1 I_{th}$, i.e., slightly above threshold. Except for the 153 K spectrum the emission is single-mode with a side-mode suppression ratio larger than 30 dB, tuning from $2029.38 cm^{-1}$ ($4.93 \mu m$) at 80 K to $1966.58 cm^{-1}$ ($5.08 \mu m$) at 194 K.

Assuming that no heating of the device occurs in pulsed-mode (100 ns pulse width, 5 kHz repetition rate), the true temperature of the active region $T_{act}$ can be estimated by comparing the emission spectra in pulsed and CW operation. The analysis shows that $T_{act}$ increases rapidly for heat-sink temperatures close to the maximum CW operating temperature due to the limited heat dissipation within the device. At the maximum heat-sink temperature $T_{hs} = 194 K$ we estimate $T_{act} = (298 \pm 10) K$, which is close to the maximum pulsed-mode operating temperature. For the thermal resistance $R_{th}$ defined as

$$R_{th} = \frac{T_{act} - T_{hs}}{P_{el} - P_{opt}}$$

we obtain with the injected electrical power $P_{el} = 2.7 W$ and the emitted optical power $P_{opt} = 1 mW$ a value of $R_{th} = (38 \pm 4) K/W$.

The CW operation performance of our devices can be further enhanced by improved thermal management to reduce the thermal resistance (e.g., by lateral overgrowth of InP increasing the lateral heat dissipation [8,20]) and by employing modified active region designs aiming at reducing the threshold...
current density (e.g., by using four quantum well active regions with double-
phonon relaxation and AlAs blocking barriers as shown in Fig. 4 and [18]).

4 Relative Intensity Noise

In this section we present first experimental investigations of the intensity noise properties of a $16 \times 2000 \mu m^2$ QC laser based on the design of Fig. 1b using lattice-matched GaInAs/AlInAs with incorporated AlAs blocking bar-
riers. The device was driven by current pulses of 100 ns length at a repetition rate of 1 kHz. The emitted light was collected by an f/1.6 mirror collima-
tor and focused onto a peltier-cooled HgCdZnTe photovoltaic detector of $\sim 150$ MHz bandwidth. After amplification using a low-noise amplifier the detected signal was split by a power divider and analyzed simultaneously by an oscilloscope and an electrical spectrum analyzer (ESA). The intensity fluctuations of the device were characterized by measuring the normalized relative intensity noise $RIN^*$ given by

$$RIN^* = \frac{(P_S - P_D)R}{B \tau_{dc} U^2},$$

where $P_S$ is the measured spectral noise power of the laser light, $P_D$ is the spectral dark noise power, and $B$ is the resolution bandwidth of the ESA. The mean detected electrical power is $\tau_{dc} U^2/R$ with the voltage of the detected optical pulse $U$, the impedance of the amplifier $R$, and the duty cycle $\tau_{dc}$.

The experimentally determined normalized relative intensity noise $RIN^*$ measured at a frequency of 9.5 MHz and a heat-sink temperature of 273 K is shown in Fig. 7 as a function of the optical power $P_{opt}$. In the investigated range of $P_{opt}$, corresponding to injection currents between $I \approx 1.08 I_{th}$ and $I \approx 1.62 I_{th}$, $RIN^*$ decreases from -99.6 dB/Hz to -115.4 dB/Hz. The experimental data can be fitted according to a simple power-law $RIN^* \propto P_{opt}^\gamma$ with $\gamma = 1.8$. For other QC lasers we found $\gamma = 1.6–1.8$. Compared to these results, in interband semiconductor lasers $\gamma = 3$ was determined in the low frequency

![Fig. 7. Normalized relative intensity noise $RIN^*$ versus optical power $P_{opt}$ of a $16 \times 2000 \mu m^2$ QC laser measured at a frequency of 9.5 MHz and a heat-sink temperature of 273 K (squares) and least square fit to the experimental data (solid line). The device is driven by current pulses of 100 ns length at a repetition rate of 1 kHz.](image)
limit and for small values of $P_{\text{opt}}$ [21]. Also for lead-chalcogenide diode lasers [22] and for vertical-cavity surface-emitting lasers [23] the same value $\gamma = 3$ was obtained.

Theoretical noise considerations for interband semiconductor lasers according to a small signal rate equation analysis, assuming single-mode operation, results in $\text{RIN}^* = C(\beta, n_{\text{sp}}, \tau_{\text{ph}})P_{\text{opt}}^{-3}$ [24], where the constant $C$ contains the spontaneous emission coefficient $\beta$, the population inversion factor $n_{\text{sp}}$ and the photon lifetime $\tau_{\text{ph}}$. Thus, we attribute our experimental finding of $\text{RIN}^* \propto P_{\text{opt}}^{-\gamma}$ with $\gamma \approx 1.8$ for QC lasers to a pump power dependence of $\beta$ or $n_{\text{sp}}$ or to multi-mode emission. This interesting observation deserves further detailed analysis in order to get a complete understanding of the intensity noise properties of QC lasers.

5 Trace-Gas Detection Using Quantum Cascade Lasers in Continuous-Wave Operation

Current scientific topics of atmospheric chemistry pose great demands on trace-gas measurement techniques based on infrared spectroscopy. The instruments should be compact to be used on – often space restricted – airborne platforms, reliable under harsh environmental and electromagnetic conditions, and easy to operate. However, the most important requirements are selectivity (no cross interference to other chemical species), versatility (several components detectable with the same technique), and sensitivity (typically within the range of detectable optical density of $10^{-6}$). For a detailed discussion of the field of infrared spectroscopy in atmospheric chemistry, the reader is referenced to [25]. Tunable diode laser absorption spectroscopy (TD-LAS) based on lead-chalcogenide diode lasers is able to fulfill these requirements with respect to selectivity, versatility and sensitivity. However, the lead-chalcogenide material technology is not mature enough to enable a more widespread application of this spectroscopic technique. The major obstacle in the further development of TDLAS is that individual lead-chalcogenide lasers are not adequately comparable in their operation characteristics. Consequently, it is currently not possible to run such a spectrometer by an untrained operator. QC lasers as a new type of a narrow bandwidth, high power infrared light source offer the opportunity to exploit the full potential of TD-LAS.

Although room temperature CW operation of QC lasers emitting at 9.1 $\mu$m wavelength was reported recently [8], most state-of-the-art QC lasers operated in CW-mode still require cryogenic cooling. For this reason several different techniques were developed to fulfill the above mentioned criteria employing QC lasers operated in pulsed-mode [26,27,28,29]. However, these techniques suffer either from limited sensitivity or from the need of high detector bandwidth to resolve the laser’s fast frequency chirp due to ohmic heating within the individual pulses, which is likely to fail under harsh elec-
tromagnetic conditions e.g., within airplanes. Therefore, we employ QC lasers operating in CW-mode using cryogenic cooling already established for lead-chalcogenide lasers. An application of atmospheric trace-gas detection already indicating the outstanding performance of QC lasers is described in [30].

At the Max-Planck-Institute for Chemistry a comparison trace-gas sensing experiment employing a lead-chalcogenide diode laser (double-heterostructure laser, Laser Components GmbH, Olching, Germany) and one of the present QC lasers (12 × 1000 µm² device with HR-coated facets of the same design used for the CW experiments presented in Sect. 3) operated in a two-laser spectrometer was designed. The optical set-up is described in [31], the applied electronics and the principles of the gas supply setup in [32]. Shortly summarized, a custom-built liquid nitrogen cryostat houses both lasers and two detectors, an InSb photodiode for the measurement and a HgCdTe photodiode for reference signal detection (both Kolmar Technologies Inc., Newburyport, MA, USA). The beams of both lasers are combined using a semi-transparent coated CaF₂ window with 50 % reflectivity and are guided into a 36 m multipass cell of Herriott type (44 hPa cell pressure) with astigmatic mirrors [33]. A small fraction of the beam is separated behind the cell to generate a reference signal for absorption line identification and locking. The emission of both lasers is guided over the absorption feature by small current ramps (about 5 mA for both lasers)¹ of 82 ms duration. The time-multiplexed operation (4.8 s integration time for every laser) was maintained by consecutive blocking of the individual beams with small chopping units. For increased sensitivity a 2f wavelength modulation (5 kHz modulation frequency) detection scheme is used [25,32,34].

Both the QC laser and the lead-chalcogenide laser were tuned to measure the P(25) absorption line of the fundamental CO band at 2037.025 cm⁻¹ (band center at about 2170 cm⁻¹). Its line strength is about a factor of 100 lower compared to the strongest CO absorptions within this band. However, it allows a small signal experiment in combination with the easy gas handling characteristics of CO. Both lasers showed single mode emission at their operating conditions near their respective threshold currents (I ≈ 1.2 Ith for both lasers) as indicated by a monochromator, as well as by their noise characteristics and étalon tuning behavior. They both show multi-mode operation at higher injection current, which typically inhibits sensitive measurements. The operating temperature of the QC laser was approximately 100 K, whereas the lead-chalcogenide laser was operated at about 82 K. At the respective operating condition the optical power emitted by the QC laser is a factor of 40 higher, as measured by a pyroelectric detector.

Figure 8 depicts a 20 min time series of calibration gas that contains 3.47 ppmv CO (ppmv = “parts per million of volume”, i.e., 1 ppmv = 1 µmol CO/1 mol air). The instrument was calibrated immediately before the the

¹ Preliminary tests showed a current tuning rate of 0.6 GHz/mA for the QC laser, which corresponds to typical values obtained for lead-chalcogenide lasers.
measurements were taken with the same gas. The concentration corresponds to an optical density at the line center of $7.29 \times 10^{-3}$. Both lasers indicate identical concentrations to within the measurement errors during the first few minutes, but small changes of the observation conditions (combined effects of laser as well as optical instabilities) lead to systematic drifts after about 10:44. Usually these instabilities are addressed with a new calibration or background measurement of the instrument by flushing it with calibration or zero gas (gas not containing the species of interest). However, the drift tends to be smaller for the QC laser, which could increase the time interval within which a recalibration is needed. We think that the higher stability has two possible reasons: (1) the considerable lower temperature tuning rate of the QC laser (the measured value of 2.2 GHz/K is a factor of 50 smaller compared to typical lead-chalcogenide lasers), and (2) the higher quality of the beam profile \[35\]. However, these effects have to be investigated in more detail in the future. The noise (precision) of the lead-chalcogenide laser measurements of about 44 ppbv (1σ; ppbv = “parts per billion of volume”, i.e., 1 ppbv = 1 nmol CO / 1 mol air) is mainly caused by detector noise due to the low laser power. The noise (precision) of the QC laser measurement is about 13 ppbv (1σ), corresponding to a detectable optical density of $2.76 \times 10^{-5}$ at an observation bandwidth of 5.7 Hz or a normalized detectable optical density$^2$ of $3.21 \times 10^{-7}$ Hz$^{-1/2}$ m$^{-1}$ (1σ). This value is a factor of four smaller compared to the value obtained for the lead-chalcogenide laser. Our present

$^2$ The relative precision of the QC laser measurements (0.34 %) is already high. Therefore, it is not excluded that a considerable fraction of the estimated noise is related to other sources in the spectrometer as e.g., pressure variations in the measurement cell or temperature uncertainties. The experiments will therefore be repeated at a lower concentration level. It is further intended to make ambient air measurement comparisons in the near future.
results are summarized in Table 1. They confirm the findings of Webster et al. who have also observed considerably improved performance of a QC laser compared to a lead-chalcogenide diode laser in the 8 μm wavelength range [30]. Webster et al. also describe an improved temperature cycling behavior from which we might anticipate, together with the lower tuning rates, an improved spectrometer handling.

Table 1. Characteristics of the time series shown in Fig. 8. A running mean over approximately 3 min is used to separate drifts from noise (precision). The former is numerically differentiated and smoothed afterwards by the same running mean to estimate drift rates. Faster fluctuations are interpreted as noise, which is given as 1σ values. Since drifts can be effectively eliminated by regular background and calibration measurements, the noise is interpreted as detectable optical density (OD), which is also given normalized to an observation bandwidth of 1 Hz and standard absorption path of 1 m

<table>
<thead>
<tr>
<th></th>
<th>unit</th>
<th>QC laser</th>
<th>lead-chalcogenide laser</th>
</tr>
</thead>
<tbody>
<tr>
<td>noise (precision)</td>
<td>ppbv</td>
<td>13.1</td>
<td>43.6</td>
</tr>
<tr>
<td>detectable OD</td>
<td>—</td>
<td>2.76 × 10⁻⁵</td>
<td>9.15 × 10⁻⁵</td>
</tr>
<tr>
<td>normalized detectable OD</td>
<td>Hz⁻¹/² m⁻¹</td>
<td>3.21 × 10⁻⁷</td>
<td>10.6 × 10⁻⁷</td>
</tr>
<tr>
<td>drift rate average</td>
<td>ppbv s⁻¹</td>
<td>0.102</td>
<td>-0.262</td>
</tr>
<tr>
<td>abs. maximum</td>
<td>ppbv s⁻¹</td>
<td>0.531</td>
<td>1.45</td>
</tr>
</tbody>
</table>

6 Summary

In summary, different designs of QC lasers emitting in the 5 μm wavelength range aiming at enhancing population inversion were presented. In a first approach, AlAs blocking barriers together with strain-compensating InAs layers were incorporated into the active regions of QC lasers using lattice-matched GaInAs/AlInAs. Doing so the confinement of electrons in the initial state of the laser transition was enhanced by selectively blocking their direct tunneling/emission into the next active region/continuum. The device performance was further improved by employing strain-compensated active regions and injectors giving rise to an increased conduction band offset. Designs based on single-phonon relaxation as published in [19] and on double-phonon relaxation [18] in combination with AlAs blocking barriers were employed. Next, the normalized relative intensity noise RIN* was investigated. For QC lasers RIN* was found to decrease more slowly with increasing optical power compared to data obtained for interband semiconductor lasers. Finally, a comparison trace-gas sensing experiment employing one of the present QC lasers
and a lead-chalcogenide laser was presented. Detecting the P(25) absorption line of CO, higher stability was obtained using a QC laser.

Acknowledgements

The authors would like to thank J. Schaub and N. Rollbühler for material growth, J. Schleife, K. Schwarz and R. Moritz for technical support, M. Mikulla, J. Wagner and G. Weimann for continuous support. Funding of the present work by the German Federal Ministry of Education and Research (BMBF) within the project “QUANKAS” is gratefully acknowledged.

References