High-Performance GaAs/AlAs Terahertz Quantum-Cascade Lasers For Spectroscopic Applications

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Abstract—We have developed terahertz (THz) quantum-cascade lasers (QCLs) based on GaAs/AlAs heterostructures for application-defined emission frequencies between 3.4 and 5.0 THz. Due to their narrow line width and rather large intrinsic tuning range, these THz QCLs can be used as local oscillators in airborne or satellite-based astronomical instruments or as radiation sources for high-resolution absorption spectroscopy, which is expected to allow for a quantitative determination of the density of atoms and ions in plasma processes. The GaAs/AlAs THz QCLs can be operated in mechanical cryocoolers and even in miniature cryocoolers due to the comparatively high wall-plug efficiency of around 0.2% and typical current densities below 500 A/cm². These lasers emit output powers of more than 1 mW at operating temperatures up to about 70 K, which is sufficient for most of the abovementioned applications.

Index Terms—Quantum-cascade laser (QCLs), terahertz (THz) spectroscopy.

I. INTRODUCTION

The invention of quantum-cascade lasers (QCLs) about 25 years ago [1] opened the path to a variety of spectroscopic approaches in the mid- to far-infrared spectral region. In particular, QCLs for the terahertz (THz) spectral region [2] allow for high-resolution spectroscopy of molecules, atoms, and ions utilizing rotational or fine-structure transitions. During the last decade, THz QCLs have been developed for the use as local oscillators in heterodyne receivers for astronomy [3]–[5]. Since 2014, a THz QCL developed at the Paul-Drude-Institut has been employed as the local oscillator on board of Stratospheric Observatory For Infrared Astronomy (SOFIA) for the detection of interstellar atomic oxygen [6]. In atmospheric science, the rotational transition of OH at 3.55 THz and the fine-structure line of atomic oxygen (OI) at 4.75 THz are of particular interest. Both can be measured with QCL-based heterodyne receivers.

Due to their high emission powers and narrow line widths in continuous-wave (cw) operation, THz QCLs are excellent radiation sources for high-resolution spectroscopy. For such applications, they have to emit radiation at a well-defined frequency, but also have to exhibit an intrinsic tuning range of 5–10 GHz. Such a tuning range is necessary in order to analyze the line shape of an absorption line, e.g., for the quantitative determination of the atom and ion densities in plasma processes. Furthermore, QCLs emitting in the atmospheric windows around 3.43, 4.32, and 4.92 THz are of interest for applications such as THz spectroscopy under pulsed megagauss magnetic fields at high-magnetic-field facilities, if the THz radiation has to be transmitted through air over a distance of about 10 m into the magnet inside a Faraday cage.

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Second, QCLs are the source of choice for many applications if a cw output power of at least 1 mW is necessary. This output power has to be correlated with a maximum operating temperature. Hence, we define a \textit{practical operating temperature} $T_{\text{pr}}$ as the temperature at which the QCL exhibits an output power of at least 1 mW emitted in a fundamental Gaussian mode.

For a near-Gaussian beam profile, QCLs with so-called surface plasmon waveguides [2] are used. However, they require designs with rather large gain in order to compensate for the lower mode confinement factor compared to the one for metal-metal waveguides [8]. In particular, for cw operation, lasers with a high gain and a low electrical pumping power, i.e., lasers with a high wall-plug efficiency, are preferred. Here, so-called hybrid designs, in which a bound-to-continuum transition is combined with direct carrier injection or resonant population of injector levels assisted by longitudinal optical phonon emission [9], have proven to be of advantage [5].

In this article, we present THz QCLs based on GaAs/AlAs heterostructures with emission frequencies between 3.4 and 5.0 THz based on the hybrid design, substantially extending the accessible spectral range of lasers based on this materials system, which has been so far reported only for 4.75 THz. Although the growth remains challenging, we have realized lasers from 11 different wafers. Optimized lasers exhibit competitive wall-plug efficiencies and rather high practical operating temperatures in cw operation. Finally, we demonstrate the operation of these THz QCLs in a mechanical cryocooler (Ricor K535) or a miniature cryocooler (AIM SL400).

\section{Designs}

For the development of the designs, we started from a laser structure operating at 4.75 THz (sample B in [10]), followed by a gradual scaling of the layer structure toward lower or higher frequencies. The QCL structures consist of 78 periods for frequencies smaller than or equal to 3.90 THz and 88 periods for frequencies larger than 3.90 THz using in all cases 8 quantum wells in each period. The corresponding frequencies of the gain maxima are achieved by an appropriate adjustment of the quantum well thicknesses and a corresponding fine-tuning of the thicknesses of some particular barriers. The quantum well, which contains the transition resonant to the energy of the longitudinal optical phonon, is Si doped with a density of up to $2 \times 10^{17}$ cm$^{-3}$. Fig. 1(a) and (b) depicts the calculated subband structures using the nominal layer thicknesses of the designs for 4.75 THz and 3.50 THz, respectively, as examples, which demonstrates that the scaling maintains the essential subband structure. Similar designs have been demonstrated by Köhler \textit{et al.} [11] and Scalari \textit{et al.} [12] for the GaAs/Al$_{0.15}$Ga$_{0.85}$As materials system with significantly lower doping levels.

In addition to the hybrid character of the design, i.e., the combination of a \textit{bound-to-continuum} laser transition with an efficient carrier extraction from the quasi-miniband and a resonant population of the injector levels utilizing scattering by longitudinal optical phonons, a unique feature of the present design compared to other recent hybrid designs such as discussed by Amanti \textit{et al.} [13] is an undoped \textit{injector} quantum well between the doped quantum well and the quantum well, in which the lasing transitions takes place, as shown in Fig. 1. Due to the applied electric field, the positive space charge in the doped quantum well and the negative space charge in the undoped injector quantum well lead to the formation of a local dipole, i.e., to local electric-field domains [14], [15]. This local dipole is expected to stabilize the laser operation over a wider range of applied field strengths by supporting the self-adjustment of the states involved in the carrier injection into the upper laser level. The self-adjustment can be explained by a concurrent reduction of the electron population in the undoped injector quantum well, since a higher applied field strength results in a stronger coupling of injector and upper laser states. While the former process leads to a lowering of the energy of the injector state, it compensates for the latter one, resulting in an extended dynamic range. This allows for a reasonable intrinsic tuning

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Conduction band profiles, subband structures, and positions of the Si doping of QCLs for (a) 4.75 and (b) 3.50 THz. The blue lines depict the laser states (11, 10), while the red lines indicate the initial (s1, s2) and final states (s0) for the main transitions, which are resonant to the energy of the longitudinal optical phonon. The thick black line presents the injector state (i). Note that the dipole matrix elements $D_{11-10}$ for the lasing transition may vary with increasing field strengths as the coupling of the lower laser level to the quasi-miniband varies while the energy separation $E_{11-10}$ is rather constant due to the vertical character of the transition. In order to evaluate the designs, gain spectra have to be calculated for a larger field strength range, as shown in [7]. For the given field strengths, the key design parameters are $E_{11-10} = 20.1$ meV, $D_{11-10} = 4.1$ nm, $E_{11-11} = 1.4$ meV, $E_{s2-s0} = 0.9$ meV, $E_{s1-s0} = 34.0$ meV, $E_{s1-s0} = 29.7$ meV with a quasi-miniband width of 21.1 meV and $E_{11-10} = 14.5$ meV, $D_{11-10} = 1.7$ nm, $E_{11-11} = 0.9$ meV, $E_{s2-s0} = 34.0$ meV, $E_{s1-s0} = 29.7$ meV with a quasi-miniband width of 16.1 meV for (a) and (b), respectively.}
\end{figure}
range of the laser modes even for designs with a vertical laser transition as shown recently [16].

The longer periods of such hybrid designs reduce the internal electrical field strengths and consequently the leakage currents. In addition, the undoped quantum well spatially separates the dopants from the energy states of the lasing transitions reducing parasitic impurity scattering. However, these rather complex structures together with the thin AlAs barriers are challenging to simulate as well as to grow. For the development of high-performance lasers, optimization procedures are necessary, which include both, simulated and empirical data, for a number of wafers with refined layer thicknesses. In particular, the background doping density [17], which is difficult to control over a longer growth campaign, may affect the effective carrier density and, hence, the formation of the local dipole. It may consequently lead to a small but significant rearrangement of the injector and laser states.

The GaAs/AlAs materials system exhibits very high barriers, which may lead to rather large interface roughness scattering. However, the influence of this scattering process on the laser transition can be neglected, since the envelope wave functions of the laser states possess a rather small amplitude at the position of the barriers/interfaces in our design with a vertical laser transition as discussed in [7]. Due to interdiffusion, the interfaces between barriers and quantum wells show a grading rather than an abrupt transition. While the realistic band structure with graded interfaces is included in our design procedure [10], possible alloy scattering is again neglected since the alloy region is located at positions where the wave functions have a small amplitude.

III. GROWTH AND REALIZATION

The lasers were grown by molecular beam epitaxy, which is particularly challenging for the very thin AlAs barriers with 2–4 monolayer thicknesses. Our approach consists in nominal growth rates of 0.11 and 0.13 nm/s for AlAs and GaAs, respectively, leading to a minimum Al shutter opening time of 5 s for the thinnest barrier and an overall growth time of about 22 h for the whole cascade structure. The average growth rates amount to 0.13 nm/s, while fluctuations in the growth rates are below 1% due to the use of a closed-loop rate control system based on optical reflection measurements [18] for the in-situ growth control. During growth, the substrate was rotated at a speed of about 12 r/min, which is adjusted for each QCL wafer. The optimization of the lasers has already started, which can be seen by the comparatively larger powers for these frequencies. The wall-plug efficiencies for lasers at 3.50 and 4.75 THz reach values larger than 1.8 and 3.4%, respectively, i.e., for typical output powers of 1 mW, electrical power meter (Laserprobe RkP-575 RF).

IV. RESULTS

Fig. 3 exhibits a compilation of operating parameters of 21 GaAs/AlAs QCLs. For 3.50 and 4.75 THz, three different wafers were used, while for 3.92 THz the lasers were fabricated from two different wafers. For all other frequencies, only one wafer was used to fabricate lasers. All QCLs are based on the hybrid active-region design. The output power of Fabry-Pérot lasers based on single-plasmon waveguides is shown for cw operation as a function of the emission frequency. The typical ridge dimensions of these lasers are about 0.12 × 1.0 mm², and their threshold current densities vary between 100 and 300 A/cm². For 3.50 and 4.75 THz, the optimization of the lasers has already started, which can be seen by the comparatively larger powers for these frequencies. The wall-plug efficiencies for lasers at 3.50 and 4.75 THz reach values larger than 1.8 and 1.1 × 10⁻³, respectively. Power values are not corrected for window transmission losses or collection efficiency and are a lower bound by a factor of about 2 to the absolute power of the lasers.

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TABLE I
LASER PARAMETERS FOR CONTINUOUS-WAVE OPERATION

| QCL | ν_{max} (THz) | ν_{mode} (THz) | T_{po} (K) | Δν (GHz) | L (mW) | J_{th} (A/cm²) | J_{max} (A/cm²) |
|-----|---------------|---------------|------------|----------|--------|---------------|----------------|
| A   | 3.5           | 3.41 – 3.50   | 65         | 5        | 4.7    | 240           | 490            |
| B   | 4.1           | 3.94 – 3.98   | 50         | 11       | 1.6    | 190           | 460            |
| C   | 4.8           | 4.67 – 4.80   | 55         | 7        | 4.4    | 220           | 510            |

ν_{max} denotes the frequency of the calculated gain maximum, ν_{mode} the frequency range of the observed lasing modes, T_{po}, the practical operating temperature, Δν the intrinsic tuning range, L the output power, J_{th} the threshold current density, and J_{max} the current density at the maximum of the output power. The values for Δν, L, J_{th}, and J_{max} have been determined for a heat-sink temperature of 30 K.

Fig. 3. Maximum output power for cw operation as a function of the center emission frequency for 21 GaAs/AlAs QCLs (asterisks) based on the hybrid active-region design measured at a heat sink temperature of 30 K. The vertical lines indicate the target frequencies of 3.36, 3.92, and 4.75 THz for fine-structure transitions of Al, N, and O atoms/ions, respectively, and 3.55 THz for OH detection. The dashed line depicts a simulated transmission spectrum of air based on the HITRAN database for ambient conditions corresponding to the USA model, mean latitude, summer, and an optical path length of 10 m, exhibiting maxima at 3.43, 4.32, and 4.92 THz.

Fig. 4. (a) L−J−V characteristics for several operating temperatures and (b) lasing spectra for several operating temperatures and current densities of QCL A with laser ridge dimensions of 0.12 × 0.94 mm² under cw operation. The vertical solid lines indicate the target frequencies of 3.43 and 3.50 THz.

Figures 3 and 4 illustrate the performance of QCLs in a continuous-wave (cw) operation mode. The tables summarize the key parameters for QCLs emitting at various frequencies:

- **ν_{max}** (THz): The frequency of the calculated gain maximum.
- **ν_{mode}** (THz): The frequency range of the observed lasing modes.
- **T_{po}** (K): The practical operating temperature.
- **Δν** (GHz): The intrinsic tuning range.
- **L** (mW): The output power.
- **J_{th}** (A/cm²): The threshold current density.
- **J_{max}** (A/cm²): The current density at the maximum of the output power.

These parameters are crucial for understanding the capabilities and limitations of QCLs in applications such as spectroscopy and sensing. The data presented in Table I are for QCLs A, B, and C, with each having distinct performance characteristics.

The operating temperatures and current densities for QCLs A, B, and C are tabulated, showing how they can be tuned to specific frequencies within the specified ranges. The threshold current densities are notably low, allowing for efficient operation even at relatively lower power levels. The output powers also vary, with QCL A demonstrating higher outputs compared to the others.

The tables highlight the importance of parameters such as Δν and J_{max}, which are critical for achieving high-resolution spectroscopy applications. The practical operating temperatures (T_{po}) provide insight into the thermal management strategies needed for sustained operation.

The figures complement the tables by visualizing the output power as a function of frequency and the lasing spectra under different conditions. These visualizations are essential for understanding the spectral characteristics and potential applications of QCLs in the terahertz frequency range.

The text concludes with a discussion on the practical implications of the presented data, emphasizing the feasibility of operating QCLs in miniature cryocoolers and the potential for high-resolution spectroscopy and sensing applications.

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4QCL A has been fabricated from wafer PDI-M4-3322. Starting from the injection barrier, indicating AlAs and GaAs layers by bold and normal font, respectively, and denoting the Si-doped quantum well (ν_{Si} = 2.0 × 10^{11} cm⁻³) by underlining its thickness, the nominal thicknesses of the layers in nm are 0.84/32.0/0.48/16.9/0.48/13.1/0.48/11.7/0.48/10.5/0.48/9.6/0.48/20.0/0.84/19.1. The quantum well doping corresponds to an average doping of 2.9 × 10^{11} cm⁻³ and a sheet carrier density of 4.0 × 10^{11} cm⁻² per period.

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Fig. 5. (a) $L$–$J$–$V$ characteristics for several operating temperatures and (b) lasing spectra for several operating temperatures and current densities of QCL B with laser ridge dimensions of $0.12 \times 0.99 \text{ mm}^2$ under cw operation. The vertical solid line indicates the target frequency of 3.92 THz.

The $L$–$J$–$V$ characteristics as well as the lasing spectra of the least optimized QCL B,\(^5\) which emits at about 3.90 THz, are shown in Fig. 5. The output power is lower than for QCL A, although it may be sufficient for high-resolution absorption spectroscopy of the fine-structure transition of N\(^+\) ions. The tuning range at 30 K is about 11 GHz, which is rather large.

Fig. 6 displays the $L$–$J$–$V$ characteristics as well as the lasing spectra of QCL C,\(^6\) which emits at 4.745 THz. It is the most mature QCL out of this series. Even for this comparatively large emission frequency, an output power of about 4 mW is achieved at 30 K, when the laser is operated in a helium-flow cryostat for laser ridge dimensions of $0.12 \times 1.05 \text{ mm}^2$. The gain maximum is close to the target frequency. Based on this design, we fabricated lasers with an improved output power using ridge dimensions of $0.08 \times 0.90$ and $0.08 \times 0.87 \text{ mm}^2$. Operated in a mechanical cryocooler, we demonstrated $T_{\text{po}} > 70$ K, as shown in Fig. 7(a). These lasers can readily be operated in a miniature cryocooler with a cooling power of about 1 W. Fig. 7(b) shows the beam profile obtained by using a TPX lens in this configuration. Using enhanced back-facet reflection, another laser out of this series operated in the miniature cryocooler has recently been shown to provide up to 8 mW output power. This QCL emits a single mode around the rest frequency of atomic oxygen, which can be tuned from $-2.7$ to $+9.4$ GHz\,[23]. A similar laser has been tested in a spectrometer to be used for plasma diagnostics. Fig. 8 shows an absorption line of NH\(_3\) at 4.767 THz. Static fine-tuning of this laser to 4.745 THz following the procedure described by B. Röben et al.\,[24] is finally expected to allow for the detection of atomic oxygen in the plasma reactor.

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\(^5\)QCL B has been fabricated from wafer PDI-M4-3417. Starting from the injection barrier, indicating AlAs and GaAs layers by bold and normal font, respectively, and denoting the Si-doped quantum well ($n_{\text{Si}} = 1.5 \times 10^{17} \text{ cm}^{-3}$) by underlining its thickness, the nominal thicknesses of the layers in nm are: 1.12/29.9/0.56/16.1/0.42/12.8/0.42/11.3/0.42/9.6/0.42/8.8/0.42/19.1/0.84/19.1. The quantum well doping corresponds to an average doping of $2.2 \times 10^{16} \text{ cm}^{-3}$ and a sheet carrier density of $2.9 \times 10^{11} \text{ cm}^{-2}$ per period.

\(^6\)QCL C has been fabricated from wafer PDI-M4-3418. Starting from the injection barrier, indicating AlAs and GaAs layers by bold and normal font, respectively, and denoting the Si-doped quantum well ($n_{\text{Si}} = 2.0 \times 10^{17} \text{ cm}^{-3}$) by underlining its thickness, the nominal thicknesses of the layers in nm are: 1.12/27.2/0.56/15.0/0.42/12.1/0.42/10.8/0.42/9.2/0.28/8.1/0.28/7.2/1.12/0.84/19.1. The quantum well doping corresponds to an average doping of $2.9 \times 10^{18} \text{ cm}^{-3}$ and a sheet carrier density of $3.5 \times 10^{11} \text{ cm}^{-2}$ per period.
Fig. 7. (a) Maximum output power under cw operation as a function of heat-sink temperature for a laser with ridge dimensions of $0.08 \times 0.87 \text{mm}^2$ emitting at 4.75 THz operated in a Stirling cooler. The dashed line indicates the power of 1 mW as a guide to the eye. (b) Beam profile of the 4.75-THz QCL (ridge dimensions $0.08 \times 0.90 \text{mm}^2$) operated in a miniature Stirling cryocooler.

V. CONCLUSION

We have shown that THz QCLs based on GaAs/AlAs heterostructures can be designed for emission at various frequencies between 3.4 and 5.0 THz with output powers of several mW using single-plasmon waveguides. We expect that QCLs emitting at any frequency in the range between 3.4 and 5.0 THz can be developed by a straightforward interpolation of the presented designs. In particular, QCLs emitting at the target frequencies indicated in Fig. 3 can be realized by relying on the basic design in the GaAs/AlAs materials system. By optimizing the balance between the electric-field-dependent gain spectra and the non-linear transport properties, i.e., the onset of negative differential conductance, the operating parameters such as the output power and $T_{\text{po}}$ may be further improved. The straightforward design of the resonators allows for a similarly straightforward fine-tuning of the laser modes so that these lasers are suitable for several practical applications in the field of high-resolution THz spectroscopy. The improved wall-plug efficiencies of GaAs/AlAs THz QCLs make them suitable for spaceborne applications, where a high $T_{\text{po}}$ is required, in particular for passive cooling.

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