Terahertz quantum-cascade lasers for high-resolution spectroscopy of sharp absorption lines

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Terahertz (THz) quantum-cascade lasers (QCLs) are currently unparalleled for high-resolution spectroscopy of very sharp absorption lines (linewidths below 100 MHz) in the range between 2 and 5.4 THz. Since the frequency range accessible by a single QCL is determined by its typically very limited tuning range, a particular QCL has to be fabricated for each specific application. We quantitatively analyze the frequencies of the modes in THz QCLs with a Fabry-Pérot resonator as a function of its length taking into account waveguide dispersion. Based on these results, we develop a process based on mechanical polishing of the front facet to adjust the emission frequency with a precision of 1 GHz. The demonstrated process makes it possible to reliably fabricate THz QCLs for the spectroscopy of very sharp absorption lines.

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I. INTRODUCTION

High-resolution absorption spectroscopy in the terahertz (THz) spectral region between 0.3 and 10 THz provides access to the energy levels of molecules and atoms due to rotational modes and fine structure splitting, respectively, which is important for technological and scientific applications. THz quantum-cascade lasers (QCLs) in continuous-wave operation are particularly well suited for this purpose, because they achieve optical output powers above 1 mW and linewidths in the MHz range.\textsuperscript{1–3} More recently, even sub-MHz frequency stabilization has been achieved.\textsuperscript{4–6} Although THz QCLs currently operate in continuous-wave mode only below 130 K,\textsuperscript{7} the availability of powerful, yet compact and portable cryocoolers greatly facilitates practical applications.\textsuperscript{8,9}

A remarkable feature of THz QCLs is that the emission frequencies (more precisely the center frequency of the gain profiles) can be selected between 1.2 and 5.4 THz,\textsuperscript{10,11} since the operation of QCLs relies on quantum-confined states in semiconductor heterostructures, which can be manipulated through bandstructure engineering. However, the exact emission frequencies are determined by the dimensions of the resonator and the waveguide dispersion. Finding the correct dimensions to reach an application-specific target frequency proved to be highly challenging especially for rather complex resonators,\textsuperscript{12} because the intrinsic tuning is usually insufficient to compensate for the uncertainty in the optical resonator length. While several methods to increase the tuning range have been proposed, they currently suffer from drawbacks such as a reduced output power, a lack of compatibility with narrow-linewidth operation, or a high complexity.\textsuperscript{13–17} In this work, we quantitatively investigate the impact of the resonator length on the emission spectrum and propose a reliable method to reproducibly obtain emission at a specific target frequency with sufficient precision. An important advantage of this method based on mechanical polishing of the front facet is that it requires only a minimal intrinsic tunability of 1 GHz due to the high precision of the polishing technique. QCLs with a small tuning range often show a higher frequency stability, enabling high-resolution spectroscopy.

While there are several different resonator types implemented with THz QCLs such as microdisk,\textsuperscript{18} ring,\textsuperscript{19} or photonic-crystal\textsuperscript{20} resonators, the ridge geometry is usually preferred for applications due to its simplicity. Since powerful anti-reflection coatings are still lacking in the THz region, the facets of the QCL chip in the longitudinal direction almost always
provide significant feedback so that a Fabry-Pérot resonator is formed. In order to suppress multi-mode operation, frequency-selecting structures such as a distributed-feedback grating\textsuperscript{21–23}, a two-section resonator,\textsuperscript{24–26} or engineered defect sites\textsuperscript{27} may be employed.

While the resonator provides feedback in the longitudinal direction, the waveguide structures confine the generated light in the lateral directions. Two different types of waveguides—the metal-metal\textsuperscript{28} or the single-plasmon\textsuperscript{29} waveguide—are usually implemented with THz QCLs and are competing due to their complementary advantages and drawbacks. The single-plasmon waveguide routinely exhibits a substantially better emission pattern due to the larger cross-section of the waveguide mode at the expense of a lower confinement factor as the mode penetrates into the substrate. The challenge in obtaining a sufficiently small threshold current for continuous-wave operation and a good power conversion efficiency with single-plasmon waveguides can be met by employing a hybrid design based on the GaAs/(Al,Ga)As\textsuperscript{30} or the GaAs/AlAs\textsuperscript{31} material system. In this work, we will focus on the single-plasmon waveguide.

In section II, the parameters determining the frequencies of the Fabry-Pérot modes are investigated numerically and experimentally. These results are the basis for the post-processing method described in section III to precisely adjust the emission frequency, which is realized by polishing the front facet of the QCL. A summary is given in section IV.

II. MODES OF A THZ QCL WITH A FABRY-PÉROT RESONATOR

A. Numerical and experimental methods

The modes of a Fabry-Pérot resonator of length $L$ obey a standing-wave condition so that their frequencies $\nu_M$ are given by

$$\nu_M = \frac{Mc}{2n_{\text{eff}}(\nu_M)L},$$

where $M$ denotes the order of the mode, $c$ the speed of light in vacuum, and $n_{\text{eff}}$ the effective refractive index of the waveguide mode. The order $M$ of the mode corresponds to the number of intensity maxima in the resonator. Note that $n_{\text{eff}}$ is frequency dependent, since we include material and waveguide dispersion in our consideration. Consequently, Eq. (1) provides an implicit function for $\nu_M$ that is determined numerically.
FIG. 1. Values for the effective refractive index $n_{\text{eff}}$ of the waveguide for ridge widths of $W = 120$ and $200 \, \mu m$ as well as for the effective group refractive index $n_{g}$ for both ridge widths. The inset shows a schematic of the single-plasmon waveguide with a width $W$. A Fabry-Pérot resonator of length $L$ is obtained by cleaving the lithographically patterned wafer pieces into chips.

Employing

$$\frac{\partial}{\partial \nu_M} [\nu_M n_{\text{eff}}(\nu_M)] \Delta \nu_M = [n_{\text{eff}}(\nu_M) + \frac{\partial n_{\text{eff}}}{\partial \nu_M} \nu_M] \Delta \nu_M = n_{g}(\nu_M) \Delta \nu_M ,$$

an approximate equation can be derived for the spacing $\Delta \nu_M$ between two adjacent Fabry-Pérot modes of order $M$ and $M + 1$ ($\Delta M = 1$)

$$\Delta \nu_M = \frac{c}{2n_{g}(\nu_M)L} ,$$

where the group effective refractive index $n_{g}$ of the waveguide mode is used.

The function $n_{\text{eff}}(\nu)$ required to calculate $\nu_M$ and $\Delta \nu_M$ is determined via a simulation of the waveguide mode employing the commercial software package JCMsuite (JCMwave GmbH), which implements the rigorous solution of Maxwell’s equations using the finite-element method. In order to take the material dispersion into account, the frequency-dependent values for the dielectric function are taken for a fixed temperature of 100 K, below which the dielectric functions change only marginally, from the literature.$^{22,32-34}$

Figure 1 shows the effective refractive index $n_{\text{eff}}$ for a single-plasmon waveguide with a width of $W = 120 \, \mu m$ and the corresponding effective group refractive index $n_{g}$ as a
The values for $n_g$ are significantly larger than the values for $n_{\text{eff}}$ throughout the investigated frequency range. Since the differences of $n_{\text{eff}}$ and $n_g$ for $W = 120$ and 200 µm are negligible, we focus on the influence of the resonator length $L$ in this work.

For the experimental determination of the emission spectrum, the THz QCL is mounted either in a mechanical cryocooler (Stirling cooler, Ricor K535) or a helium flow cryostat (Oxford Instruments Optistat-CF-V) to be operated at heat sink temperatures of 35 or 40 K. The emission spectrum is measured with a high-resolution Fourier transform spectrometer (Bruker IFS 120HR) operated with a spectral resolution of 0.3 GHz. For continuous-wave operation, the injection current is provided by a source measure unit (Keithley 2635A). For pulsed operation, a current pulse source (Avtech AV-107C-B) providing a pulse width of 1 µs at a repetition frequency of 50 kHz is used. Pulsed operation is less sensitive to variations in terms of the thermal resistance at interfaces such as between the cold finger and the submount. These variations typically occur between alternating mounting/unmounting steps and lead to different chip temperatures for the same cold-finger temperature in continuous-wave operation. Thus, pulsed operation is employed if quantitative reproducibility is required. The selected QCL,\textsuperscript{35} which was processed into a single-plasmon waveguide with a width of $W = 120$ µm through photolithography and wet etching using H$_2$SO$_4$:H$_2$O$_2$:H$_2$O (1:1:8) chemistry, emits around 4.7 THz. The Fabry-Pérot resonator obtained by cleavage had a length $L$ of 1,585 µm as determined with an optical microscope.

B. Analysis of the modes in Fabry-Pérot resonators

Figure 2(a) shows the emission spectrum of the 4.7-THz QCL for four different current densities. Due to intrinsic tuning induced by current changes,\textsuperscript{36} a red shift of 4.2 GHz of the mode with the largest frequency is observed. Furthermore, the calculated and measured mode positions agree fairly well. The spacing between the different modes for 336 and 368 A cm$^{-2}$ are equal within the resolution of the spectrometer (0.3 GHz) and amounts to 20.3 GHz. The spacing of the calculated modes is somewhat larger and amounts to 22.4 GHz, since the simulation of the waveguide mode performed to determine $n_{\text{eff}}(\nu_M)$ and $n_g(\nu_M)$ did not include cavity pulling effects.\textsuperscript{36}

Figure 2(b) shows a comparison of the calculated modes for the original QCL ($\Delta L = 0$ µm) with the results of a corresponding calculation for two additional resonator lengths.
FIG. 2. (a) Emission spectra for the employed QCL for four different current densities between 231 and 368 A cm$^{-2}$. The QCL was operated in continuous-wave mode in the helium flow cryostat at 35 K. The dashed lines indicate the frequencies of the modes as calculated from Eq. (1). (b) Comparison of the calculated modes of the employed QCL and the results of a corresponding calculation for resonator lengths of $L = 1585 \, \mu m + \Delta L$ that are smaller by 1 and 2 $\mu m$ ($\Delta L = -1$ and $-2 \, \mu m$). As expected, we observe a blue shift of the Fabry-Pérot modes for shorter resonators, which amounts to 2.6 or 5.2 GHz for a resonator length reduced by 1 or 2 $\mu m$, respectively. This example obtained through a rigorous calculation based on Eq. (1) results in a proportional dependence between $\Delta L$ and the frequency shift $\Delta \nu_L$ for small values of $\Delta L$ as derived from Eq. (1)

$$\Delta \nu_L = -\frac{M_c}{2n_g(\nu_L)L^2} \Delta L.$$ (4)

Although these results clearly show that the emission frequency of the Fabry-Pérot modes can be controlled by means of the resonator length, achieving the required precision is very demanding. In our case, for instance, a precision of $\pm 1 \, \mu m$ would lead to an uncertainty in the emission frequency of $\pm 2.6$ GHz as can be readily inferred from Fig. 2(b), requiring a tuning range of 5.2 GHz to compensate. However, intrinsic tuning mediated by the injection current amounts only to 4.2 GHz in this case. Consequently, a precision of less than $\pm 1 \, \mu m$ for the definition of the resonator length is required.
III. PRECISE ADJUSTMENT OF THE RESONATOR LENGTH

A. Approach

The Fabry-Pérot resonator is formed by the longitudinal facets of the QCL chips, which are usually obtained by cleavage. Unfortunately, even dedicated accessories or machines for cleaving have typical precisions well above 1 µm, which is insufficient for the desired control over the frequencies of the Fabry-Pérot modes. Employing an approach different from cleaving to define the resonator length such as lithography-based methods or focused ion beam milling is not readily possible with single-plasmon waveguides, since the waveguide mode penetrates several tens of µm into the substrate and extends over almost the entire width of the stripe amounting to 120 µm. Therefore, our approach is to precisely adjust the resonator length after cleavage. We realize this post-processing step by means of mechanical polishing of the front facet. The compatibility of our approach to a fully operational QCL enables us to perform several consecutive alternating polishing and measurement runs with the same QCL chip.

B. Polishing technique

In order to measure the emission spectrum prior to polishing, we mounted and wire-bonded the QCL so that it is fully operational. The mounting of the QCL was performed with a small overhang of the front facet over the submount edge of less than 100 µm so that the polishing process is facilitated while the thermal coupling to the submount is still sufficient. The QCL was fixed on a dedicated holder for polishing using a Buehler EcoMet 250 polishing machine. The housing of our cryocooler was adapted to receive and cool the entire polishing holder with the QCL still mounted, which enables us to measure the emission spectrum after several consecutive polishing runs without removing the QCL from the polishing holder. This approach avoids any additional alignment error of the QCL facet with respect to the polishing plane. The somewhat reduced thermal coupling due to the additional thermal interface can be neglected since the QCL is operated in pulsed mode for the measurement of the spectra after each polishing run. The correspondence between the frequencies of the modes in pulsed operation and the ones in continuous-wave operation is determined before polishing.
We found a diamond lapping film with a grain size of either 0.1 or 0.5 \( \mu m \) operated with a rotation speed of 30 and 10 revolutions per minute (rpm), respectively, best suited for a precise removal of the material from the facet. It turned out that the rate of material removal fluctuates between subsequent polishing runs so that the polishing time as a parameter is insufficient to control the amount of removed material. To improve the control over the polishing process, we developed a method that we refer to as *alternating-grain-size technique*. For illustrative purposes, Fig. 3(a) shows an optical micrograph of part of a chip facet, which has first been polished with a grain size of 0.1 \( \mu m \) for about 50 s and afterwards with a grain size of 0.5 \( \mu m \) for about 20 s. The polishing always starts in the upper left corner and progresses toward the opposite corner in the direction indicated by the arrow as a result of the rotation direction and sample orientation. The dashed line indicates the boundary to which the polishing with the coarse grain size of 0.5 \( \mu m \) has already progressed. It therefore separates the region on the chip facet with a roughness corresponding to the grain size of 0.5 \( \mu m \) and the one corresponding to a grain size of 0.1 \( \mu m \). The degree of roughness is clearly distinguishable in the optical micrograph. The monitoring of the

FIG. 3. (a) Optical micrograph of part of a polished chip facet for the illustration of the alternating-grain-size technique. The facet has been first polished with a grain size of 0.1 \( \mu m \) and afterwards with a grain size of 0.5 \( \mu m \). The polishing is a gradual process starting in the upper left corner and progressing in the direction of the arrow. The dashed line indicates the boundary between the area to which the polishing process with a grain size of 0.5 \( \mu m \) has progressed and the area which shows the smaller roughness of the previous polishing process carried out with a grain size of 0.1 \( \mu m \). The ellipse marks the area over which the waveguide mode extends (area, which includes the QCL mesa). (b) Optical micrograph of the same area as in (a) after polishing run 6 (grain size 0.1 \( \mu m \)).
In order to investigate the reproducibility of the polishing approach, we performed six polishing runs, which are summarized in Table I. Only polishing runs 1 and 2 have been carried out without applying the alternating-grain-size technique. For polishing runs 3 and 4, the polishing was carried out until the full area shown in Fig. 3(a) was processed. For polishing runs 5 and 6, the polishing was only performed until the region marked by the ellipse in Fig. 3(a) was fully processed. This region encompasses the area occupied by the waveguide mode, which is the relevant region forming the actual “mirror” of the resonator. Polishing runs 5 and 6 represent the smallest possible steps for the reduction of the resonator length based on the grain size of 0.5 and 0.1 µm, respectively. Figure 3(b) shows the chip facet after polishing run 6, which exhibits a smaller roughness in the relevant area marked with the blue ellipse than after run 5, since a grain size of 0.1 µm has been employed.
FIG. 4. (a) Comparison of the spectra before the polishing (referred to as polishing run 0) and after polishing runs 1 to 6 as indicated. The spectra were taken with the QCL mounted onto the polishing holder and operated in pulsed mode within the Stirling cooler at a cold-finger temperature of 40 K. For each polishing run, three spectra at the indicated current densities have been measured.  

(b) Mode frequencies extracted from (a) as a function of the polishing run. (c) Incremental frequency shift of the modes after each polishing run extracted from (b). (d) Incremental reduction of the resonator length due to polishing estimated from the average values of the incremental frequency shift in (c) using Eq. (1).

C. Results

Figure 4(a) shows the emission spectra measured before polishing (polishing run 0) and after each of the six polishing runs for current densities of 260, 280, and 300 A cm$^{-2}$. Figure 4(b) displays the mode frequencies extracted from Fig. 4(a) as a function of the polishing run. The spectra for every current density and polishing run exhibit only a single mode. For polishing run 0 (before polishing), a large frequency difference appears between 280 and 300 A cm$^{-2}$, which originates from a hopping of a Fabry-Pérot mode of order $M_0 + 1$ to a mode of order $M_0$. The mode of order $M_0 + 1$ does not appear after polishing run 1, in which the length was reduced by a rather large amount. Consequently, the frequency of the mode of order $M_0 + 1$ is so far away from the gain maximum after polishing run 1 that it is fully suppressed for all current densities in favor of the mode of order $M_0$. Considering only the mode of order $M_0$, we observe a red shift of the mode due to current-induced tuning as
The current density is increased from 260 over 280 to 300 A cm\(^{-2}\). The value of the tuning amounts to about 2 GHz, which is smaller than the value previously obtained from Fig. 2 for continuous-wave operation. This trend has been recently observed for several THz QCLs.\(^{36}\)

The polishing of the front facet in the different polishing runs leads to a successive blue shift of almost 16 GHz in total as shown in Fig. 4(b). Figure 4(c) depicts the incremental frequency shift between successive polishing runs for the three current densities. While the values for the different current densities should be equal in the ideal case, they exhibit small variations due to uncertainties in the determination of the emission frequency and current density. Thus, an average value is calculated for each polishing run. The values of the incremental frequency shift correspond to specific incremental values of the material removal, i.e., of the reduction of the resonator length. Figure 4(d) shows the values of the length reduction estimated using Eq. (1).

While the length of the resonator has been reduced by about 3.3 \(\mu\)m in polishing run 1, the corresponding value for polishing run 2 is substantially smaller, although the reduced grain size in run 2 has been partly compensated by a higher rotation speed. This substantial difference illustrates that the polishing conditions fluctuate between the different polishing runs so that the monitoring by means of the alternating-grain-size technique is required. Employing this technique, the ratio of the length reduction between polishing runs 3 and 4 due to the different grain sizes is significantly smaller than the respective ratio between polishing runs 1 and 2. For polishing runs 5 and 6, the ratio of the length reduction is approximately the same as the one for polishing runs 3 and 4, which demonstrates a high degree of reproducibility. The smallest possible amount of polishing has been performed with polishing runs 5 and 6, for which a reduction of the resonator length of 0.5 and 0.2 \(\mu\)m, respectively, has been obtained, which corresponds to a frequency shift of 1.5 and 0.5 GHz, respectively.

Figure 5 shows the output power of the QCL before polishing and after the last polishing run. The values of the output power are somewhat above 3 mW in both cases and are identical within the error margins of the measurements. This result clearly shows that the polishing does not affect the output power, which is of paramount importance for the technique to be suitable for real-world applications. The polishing technique benefits from the large vacuum wavelength in the THz range, which is about two orders of magnitude larger than in the visible range. Therefore, the emitted THz radiation is not significantly affected
by the surface roughness, since the wavelength is substantially larger than the correlation length of the facet roughness.

The required precision of the polishing technique ultimately depends on the tunability of the QCL. If we assume a fixed value of 1 GHz for the tunability, which is a rather conservative assumption, we can calculate the required precision of the polishing process as a function of the emission frequency and the resonator length employing Eq. (1). Figure 6 shows the results for three different emission frequencies. The required precision is more demanding for shorter resonators and higher emission frequencies. Since the parameters employed for the demonstration of the polishing process are already in the submicrometer range, the polishing technique is expected to be applicable to other relevant emission ranges and resonator lengths.

IV. SUMMARY

In this work, we quantitatively analyzed the frequencies of the modes in THz QCLs with a Fabry-Pérot resonator as a function of its length, taking into account material and waveguide dispersion. We found that the emission frequencies depend most sensitively on the resonator length $L$ so that a control of $L$ with a precision below 1 µm is required for a frequency shift of about 1 GHz. Consequently, an intrinsic tuning range of 1 GHz is sufficient to reach

![Figure 5](http://example.com/figure5.png)

**FIG. 5.** Voltage and optical output power of the QCL before polishing (red curves) as a function of current density and after polishing run 6 (blue curves) at a heat sink temperature of 35 K operated in pulsed mode within the helium flow cryostat.
FIG. 6. Length adjustment $\Delta L$ of a resonator that induces a blue shift of a Fabry-Pérot mode by 1 GHz for initial mode frequencies of 2.5, 3.5, and 4.5 THz. The value of $\Delta L$ is thus a measure for the required precision of the polishing to reach a specific frequency assuming that the intrinsic tuning range amounts to 1 GHz.

a specific absorption line (linewidth of 100 MHz and below) and completely sweep over it so that the non-reversible nature of the tuning technique does not impede the envisaged application. We demonstrated that the required precision below 1 $\mu$m for the definition of the resonator length can be achieved by first cleaving the QCL chip, which defines the resonator length with a low precision, and subsequently adjusting the length precisely by mechanical polishing of the front facet aided by the alternating-grain-size technique. Since our approach is compatible with a fully operational QCL, the emission spectrum can be measured prior to polishing in order to determine the required correction of the resonator length. In addition, the emission spectrum can also be measured after every polishing run in order to monitor the obtained frequency shift of the modes. Finally, the alternating-grain-size technique enables the reduction of the resonator length in reproducible steps below 1 $\mu$m. This technique represents a reliable procedure to fabricate a THz QCL for the spectroscopy of a specific absorption line so that the QCL is only required to be tunable by 1 GHz.

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The QCL has been fabricated from wafer PDI-M4-2594. Starting from the layer closest to the substrate, indicating $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$ and GaAs layers by bold and normal font, respectively, and denoting the Si-doped quantum well ($n_{\text{Si}} = 8 \times 10^{16} \text{ cm}^{-3}$) by the underlined font, the nominal thicknesses of the layers are: $3.3/15.2/3.8/16.9/2.1/6.9/2.1/7.6/1.8/9.2/1.3/10.1/0.9/16.3/1.9/25.4$. This sequence was repeated 88 times, after which a final barrier with a thickness of 3.3 nm was grown.
