Physics and technology of Terahertz quantum cascade lasers

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ABSTRACT

Even though already in the seventies, right after the invention of the quantum cascade laser (QCL) concept, it was argued that this device could be operated in the THz (far-infrared) range of the electromagnetic spectrum, it was only in 2002 that the first working THz QCL was demonstrated. Soon afterwards, the progress was very rapid; in the space of 2–3 years, operating temperatures were raised, single-mode DFB devices were produced, applications as local oscillators in heterodyne transceivers were implemented, frequency coverage was extended to the whole 1–5 THz region. In the last few years, technological advancement has continued to improve performances: the maximum operating temperature has now reached about 250 K and about 1 W peak output power has been demonstrated. Several beam engineering techniques have been implemented, with the scope of enhancing spectral purity, improving beam quality and achieving vertical emission. In parallel, various approaches have been devised that allow frequency tunability of the emitted light, with the most efficient schemes achieving a tuning range of about 10% of the central emission frequency. Even the generation of frequency combs directly from THz QCLs has been obtained, by employing dispersion compensated waveguides and an intrinsic material non-linearity. This manuscript reviews the physics underlying the operation of THz QCLs, the technology developed to advance laser performances, and highlights the latest most promising progresses in this fascinating area of opto-electronics.

ARTICLE HISTORY

Received 23 October 2020
Accepted 17 February 2021

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1. THz spectral region: applications and perspectives

The terahertz (THz) spectrum, also known as far-infrared, conventionally spans the frequency range from 0.1 to 10 THz, corresponding to the wavelength interval from 30 to 3000 micron wavelength. For frequencies up to few hundred GHz, oscillators based on electronic microwave devices are nowadays largely employed in several applications. On the other side of the ‘terahertz gap’, in the near- to mid-IR region, semiconductor lasers are largely available. All these sources are based on compact, solid-state devices and easily operate at room temperature in continuous wave (CW) mode. At present, however, the widespread exploitation of several applications of THz radiation are still hampered by the lack of convenient, high-brilliance radiation sources, despite the progressively increasing interest in many strategic fields and the exponentially growing impact of THz research on the international industrial market [1,2].

One of the traditional applications of THz radiation is spectroscopy. Many chemical species have indeed very strong characteristic roto-vibrational absorption lines in the THz, with strengths $10^3$–$10^6$ times stronger than in the microwave region. Far-infrared spectroscopy was originally performed with incoherent thermal sources and Fourier-transform spectrometers using cryogenic bolometric detection. Sensitive approaches can be alternatively implemented by transmitting or reflecting tunable narrowband terahertz beams. The lack of coherent sources in this range was first filled by optically pumped fixed-frequency FIR lasers [3]. Generation of sidebands and mixing with microwaves in metal-insulator-metal (MIM) diodes provide broad spectral coverage with kHz level linewidth and accuracy [4–6], producing plenty of precise frequency measurements of atomic and molecular transitions.

Astronomy and space science have been the first major playground for THz technology [7]. As a topical example one-half of the total luminosity of the galaxies and 98% of the photons emitted since the Big Bang fall into the terahertz gap [8]. This radiation is then a natural probe of cool interstellar dust inside galaxies, and the study of the discrete lines of light molecular species can give nice insight into star formation and decay. Furthermore, THz gas spectroscopy in the stratosphere and upper troposphere is useful for the study of chemical processes related to ozone depletion, pollution monitoring, and global warming. Molecular spectroscopy applications have expanded over the years to other research and industrial contexts, including for instance identification and quantification of different chemicals in a variety of substances like pharmaceuticals [1], or even plasma fusion diagnostics [9].

Beyond THz lasers, T-Ray spectroscopy and imaging are also performed using either difference frequency generation from visible laser diodes, or ultra-short (broadband) terahertz pulses generated by rectification of optical pulses and coherently detected using methods like electro-optic sampling [1,2]. Imaging, in particular, has seen tremendous scientific advancement and
commercial interest and is performed either by rapidly scanning the sample or via free-space two-dimensional sampling on charge-coupled device (CCD) arrays [10]. THz imaging is extremely appealing because many materials that are transmissive in the terahertz are opaque at visible frequencies and vice versa. Non-invasive medical imaging of teeth or sub-dermal melanoma has been demonstrated. Monitoring of water levels in plants, fat content in packaged meats, and manufacturing defects in automotive dashboards and high voltage cables have all been performed. Due to its non-destructive nature, this method of analysis has potential for many industrial applications (aeronautical industry, mechanical industry, pharmaceutical industry) requiring high-speed data acquisition, on-line monitoring and software analysis [1,2]. Further applications include sensing of biomolecules (DNA, proteins), wireless communications, high-speed signal processing. Many security applications are also realized by means of THz radiation, like radar modeling as well as baggage and people check point screening for hidden threats [1,2].

2. Active regions of THz QCLs

Most present-day equipment available for THz generation is bulky, expensive, and often suffers from low output powers. Consequently, a new generation of compact, reliable THz sources would be the key for the development of the still mostly untapped THz range. As discussed below, THz emitting QCLs are proving to be good candidates to fulfil this task.

The first report on QCLs operating at THz frequencies, i.e. below the optical phonon energies, is relatively recent [11]. Design and fabrication of such long-wavelength QCLs was challenging: at these energies (a few meV), e-e scattering opens indirect relaxation channels; the huge free-carrier absorption ($\propto \lambda^2$) becomes a dominant factor of the waveguide losses; interface roughness and resonant features in the semiconductor dielectric constant must be taken into account, the design of optical waveguides faces practical limitations induced by the layer thickness technological feasibility, leakage channels in the transport are almost unavoidable owing to the small energy separation between the levels. And of course, it is important to stress that for intersubband transitions the spontaneous emission lifetime is typically much longer (microseconds) compared to the non-radiative lifetimes (picoseconds), so that radiative relaxation plays no role in transport below threshold.

Presently, several active region designs and their combinations have successfully been implemented. Among them, the most common ones are the chirped superlattice (CS) of the first THz QCL [11], the bound-to-continuum (BTC) [12], the resonant-phonon (RP) [13] and those based on indirect-pump injection, named scatter-assisted (SA) [14].

The CS design is based on the coupling of several quantum wells together in a superlattice to create a sequence of closely spaced levels (minibands) when the
appropriate electric field is applied. The radiative transition is designed to take place from the lowest state of the upper miniband to the top state of the lower miniband, yielding a large inter-miniband dipole matrix element for the radiative transition. Depopulation occurs mainly via electron–electron-assisted intraminiband scattering inside a wide (usually 15–20 meV) miniband (see Figure 1a). This process is heavily favored over inter-miniband scattering between the two lasing levels owing to the large number of final states available in the lowest miniband. In the first experiment, the miniband width was kept below the transition energy, in order to avoid photon re-absorption, meaning that direct LO-phonon scattering could not take place, although emission of LO-phonons was still the dominant cooling mechanism of the electron gas.

Alternative designs, based on a BTC scheme, have been proposed employing the same material system [12]. While the lower radiative state miniband-based depopulation follows the same concept as above, the upper radiative state is, in this case, a partially localized level that sits in the middle of the minigap. Due to the increased diagonal nature of the radiative transition, the BTC design experiences a reduced non-radiative scattering out of the upper state (see Figure 1b). Compared with a CS design, the oscillator strength of the transition is reduced by as much as 40% since the overlap of the upper level with the miniband states drops. The injection

Figure 1. Schematic diagram of four THz QCL active regions: (a) chirped superlattice [11], (b) bound-to-continuum (reproduced from [12] with the permission of AIP Publishing), (c) resonant-phonon [13] (reprinted by permission from Springer Nature: [22] copyright 2007), (d) scattering assisted (reproduced from [14] with the permission of AIP Publishing).
process, however, becomes more selective, as the injector states couple more strongly with the upper lasing state increasing the injection efficiency, which is advantageous for the device temperature performance.

Subsequently [13], a design exploiting the optical phonon emission for the depopulation of the lower lasing level has been demonstrated (see Figure 1c). In the RP concept, the collector and injector states are spatially separated from the transition and designed to be below the lower radiative state by approximately the longitudinal optical phonon energy, so that electrons can be rapidly funneled into them by emitting a LO-phonon. Since the lower lasing state wavefunction is spread over several quantum wells, it maintains a strong spatial overlap with the injector states, allowing LO-phonon scattering on a sub-picosecond time scale, without dramatically reducing the transition dipole.

More recently the SA design has been introduced [14]. Such scheme relies on an injection energy state, lying about one LO-phonon energy above the upper radiative state. Carriers are injected into the former by resonant tunneling from the upstream extraction level. At this electric field, the upper and lower lasing states are both decoupled from any resonant transport, allowing a high phonon-assisted injection efficiency. Moreover, this design relies on a large diagonality of the optical transition. By choosing a strong injection-extraction tunnel coupling, coherent transport and subsequent accumulation of carriers in the state with longest lifetime (the upper state) can then be achieved. The ensuing large population inversion is also linked to low carrier concentration in the short-lifetime injection/extraction states, reducing the effect of thermal backfilling of the lower radiative state.

For some time, it has been argued that the SA active region would represent the way forward to increase the maximum operating temperature of THz QCLs, thanks to the robustness of its injection efficiency with temperature. On the contrary, the last couple of years have witnessed great progress in the more conventional RP designs. The approach here has focused on reducing the number of quantum wells in each period, down to the ultimate limit of only two [15–17], in order to maximize the gain per unit length and eliminate unnecessary states whose electrons would not contribute to the amplification [17]. As discussed below, this concept has been instrumental for the highest operating temperature THz QCLs demonstrated to date.

The intersubband gain in a QCL is also strongly dependent on the material choice, since it is influenced by both electron effective masses $m^*$ in well and barriers and by the conduction band discontinuity $\Delta E_C$. In principle, any material system having a large enough band offset to allow for the intended transition energies could be used. At present, GaAs/AlGaAs heterostructures grown by MBE with Al concentration varying in the $0.05–0.25$ range have been the most successful choice for THz QCLs, mainly due to the conduction band offset values that assure accurate tailoring of the radiative transitions, to the
absence of alloy scattering, and to the high level of maturity achieved in the growth. Nevertheless, InGaAs/InAl (Ga)As/InP \cite{18}, InGaAs/GaAsSb/InP \cite{19} and InAs/AlSb/InAs \cite{20} based QCLs have been also developed with the objective of exploiting the low electron effective mass in the well material of these systems. In particular, In\textsubscript{0.53}Ga\textsubscript{0.47}As/In\textsubscript{0.52}Al\textsubscript{0.48}As/InP THz QCLs \cite{21} have achieved maximum operating temperatures of 155 K and high peak output powers of more than 0.5 W.

3. Waveguide configurations

The critical problem of designing an optical waveguide suitable for very long wavelengths has been one of the main hurdles for the development of THz QCLs. Two schemes have been successfully implemented to allow laser action across the THz. In the first one \cite{11}, named surface-plasmon (SP) waveguide, the mode is based on an electromagnetic wave arising at the interface between two media having opposite-sign dielectric constants (i.e. metal and semiconductor). The optical confinement is provided by the interplay between the SP at the top metallization and the quasi-metallic one created by the thin (50–70 nm) heavily doped buried contact. The surface-plasmon decay length is extremely short in the metal, while it decays slowly in the semiconductor, therefore extending well into the substrate. The overlap with the heavily doped region, however, is small, so that the free carrier loss is minimized (see Figure 2a), although the mode confinement factor $\Gamma$ is significantly below unity ($\Gamma = 0.2–0.5$). In addition, the facet reflectivity is almost comparable with the Fresnel value ($R \sim 0.3$) meaning that the mirror losses are in the range $\alpha_m = -(1/L)\ln R = 7.5–4.5$ cm\textsuperscript{-1} depending on the cavity length $L$. The waveguide losses are, on the contrary, much lower (3–5 cm\textsuperscript{-1}), leading to a favorable ratio of mirror to waveguide losses, yielding high output powers and efficiencies. In addition, the large mode size results in a relatively low-divergence beam (~30°).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{Optical mode intensity in a single plasmon (a) and double metal (b) THz QCL. The plot reports a cross section of a ridge waveguide geometry.}
\end{figure}
In the second scheme [22,23], named double-metal (DM) waveguide, the mode is essentially bound between a buried metal layer and a metal strip that sits on the surface of the semiconductor (see Figure 2b). This configuration allows a complete optical confinement even without the need to define an etched ridge for lateral confinement. Despite the $\Gamma \sim 1$ mode confinement factor, the fabrication of devices with adequate thermal and mechanical properties is strictly related to the necessary metal bonding to a host substrate. In particular, the careful choice of the metal sequence used may lead to a considerable improvement of the device thermal performance [24]. Nowadays In-Au, Au-Au or Cu-Cu interfaces are conventionally employed for the fabrication of DM waveguides. A careful analysis of the thermal management of THz QCLs based on different DM interfaces has been performed via micro-probe band-to-band photoluminescence experiments. If one compares SP and DM waveguides the use of DM waveguides is always beneficial to improve the device thermal management [24]. This becomes much more relevant at operating temperatures $T > 100$ K where the metal thermal conductivity is significantly higher than GaAs. Furthermore, among the various materials for DM waveguides, it is clear that the higher Cu thermal conductivity is highly preferred, decreasing by up to 40% the thermal resistance [24].

Heat extraction from THz QCLs is also hindered by the large device thermal resistance associated with the low heat conductivity of QCL active regions. The thermal conductivity of QCL active regions is largely anisotropic with the cross-plane ($k_\perp$) component much smaller than that of the bulk constituent materials [25]. The main reason is related to the increased rate of phonon scattering by interfaces in heterostructures with layer widths comparable or smaller than the phonon mean free path. Micro-probe band-to-band photoluminescence experiments have been employed to study the thermal management of THz QCLs and to extract their thermal conductivity [25]. They show that $k_\perp$ decreases exponentially as the temperature is increased.

Due to the impedance mismatch of the subwavelength mode at the ridge facet with free space propagating modes, DM waveguides exhibit enhanced facet reflectivity of $R = 0.5 - 0.9$ (depending on the dimensions) [23]. As a result, these waveguides are characterized by mirror losses ($\alpha_m = 1 - 2$ cm$^{-1}$) much smaller than the waveguide losses ($\alpha_w = 10 - 20$ cm$^{-1}$), meaning that only a small fraction (5–20%) of the generated photons escape the cavity. The scaled loss to confinement ratio $(\alpha_m + \alpha_w)/\Gamma$, which determines the lasing threshold, is better than for SP waveguides [23].

Only a small fraction of the terahertz power, however, can be collected, owing to the large beam divergence ($\sim 60^\circ$). High efficiency, low divergence, double-metal devices have been practically realized by improving the laser facet coupling using horn antennas [26–28], hyperhemispherical silicon lenses [29], on-chip hollow rectangular waveguides [30] or plasmonic collimators [31], but also extracting light from the whole device area, either through 2nd-order [32–
4. Electronic properties of THz QCLs

In unipolar optoelectronic devices, electrons release the excess energy gained from the applied electric field by phonon scattering. The equilibrium between the input power and the energy loss rate gives rise to hot-electron populations described by Fermi-Dirac distributions at temperatures significantly larger than the lattice one [43,44]. It has been well established both theoretically and experimentally [45,46] that in QCLs the electron-longitudinal phonon (LO) scattering is the dominant inter-subband scattering mechanism for subband separations greater than the LO-phonon energy $E_{LO}$. For inter-subband transitions at energies lower than $E_{LO}$, non-radiative relaxation is dominated by the combination of electron-electron (e-e) scattering, electron-impurity and interface roughness scattering, as well as by LO-phonon scattering of the high-energy tail of the subband electron distribution. However, even when devices are operated at low temperature, a non-equilibrium electron distribution may exist in the subbands. Electron scattering with ionized impurities may play an important role in intersubband transport, and, in some cases, may be stronger than e-e scattering. On the contrary, acoustic phonon inter-subband scattering (in the 100 ps range [47]) is relatively inefficient, especially at low temperatures where the phonon population is small. Calculation of interface roughness scattering is difficult since it requires the detailed modeling of the actual interface structure. Its importance strongly depends on the overlap of the wavefunctions with interfaces, as well as on growth characteristics. It may play an even more important role in intrasubband scattering where it can contribute to radiative linewidth broadening. Considering other intrasubband channels, LO-phonon scattering is an important process in cooling the subband electron gas. The effect of intrasubband e-e scattering is to thermalize the electron distribution inside a particular subband. Bi-intrasubband e-e scattering involves an electron scattering with another electron in a different subband without either electron leaving its subband. Energy is exchanged without affecting occupation. This process determines the rate at which electrons, in different subbands, reach their equilibrium temperatures.

The interplay between the above processes can lead to different temperatures $T^*_{j}$ for the electronic subbands $j$ [44]. Specifically, at electronic sheet densities $\geq 10^{11}$ cm$^{-2}$ e-e scattering is fast enough to create Boltzmann-like subband distributions characterized by electronic temperatures $T^*_{j}$ higher than the lattice one ($T_L$) at injected currents close to the laser threshold. Although subband
Figure 3. Device pictures of THz QCLs with different outcoupling mechanisms. a) Horn antenna (reproduced from [27] with the permission of AIP Publishing), b) plasmonic collimator (reprinted by permission from Springer Nature: [31] copyright 2010), c) antenna microcavity [42], d) ring extractor (reproduced from [38] with the permission of AIP Publishing).
thermalization and the related concept of subband temperature is questionable at densities $<10^9$ cm$^{-2}$, it still holds at densities of $3-5 \times 10^{10}$ cm$^{-2}$ $[48]$ and can be therefore safely applied to THz QCLs. THz QCLs are indeed ideal systems to study hot-electron populations due to the large electrical input power, the inherently high thermal-resistance $[24,25,49,50]$ and the limited e-lattice relaxation efficiency $[51,52]$. In the last few years, micro-probe photoluminescence spectroscopy $[24,25,44,49–52]$ has established as an ideal tool to experimentally assess the hot electron distributions and the thermal properties of THz QCLs based on different active region designs, offering a perspective for the inclusion of the electronic temperature in the general theory of semiconductor lasers and, specifically, THz QCLs.

Microprobe band-to-band photoluminescence (PL) allows space-resolved experiments with a 0.5 μm spatial resolution. The Kr laser excitation provides the valence band holes needed to probe the electronic population via band-to-band radiative recombination, while ensuring a good compromise between negligible electron heating and high signal intensity. If a voltage is applied to a QCL, several conduction subbands may be populated. The radiative recombination between populated conduction subbands and photogenerated holes gives origin to additional structure on the high-energy side of the PL spectra, whose intensity is well reproduced by the following expression:

$$I_{PL}(E) \propto \sum_{j=1}^{4} \sum_{k=1}^{4} A_{jk} E_{jk}^{4} |\langle \psi_{j} | \psi_{k} \rangle|^{2} \mathcal{L}(E)$$

where $A_{jk} = n_{j} \times p_{k}$ and $n_{j}$ and $p_{k}$ are the populations of the conduction (j) and valence (k) subbands. The lineshape function $L(E)$ is obtained joining a Lorentzian with a phenomenological broadening $\Gamma$ and an exponential decay on the high-energy side of the main peak, respectively:

$$\mathcal{L}(E) = \begin{cases} \frac{A}{2\pi (E - E_{jk})^{2} + \Gamma^{2}} & E < E_{jk} + \Delta_{j} \\ Ae^{-\left(\frac{E}{K_{B}T_{e}^{j}}\right)} & E > E_{jk} + \Delta_{j} \end{cases}$$

where $T_{e}^{j}$ are the electron subband temperatures, $K_{B}$ is the Boltzmann constant, $E_{jk}$ is the energy of the $j \rightarrow k$ transition, and $\Gamma$ is transition linewidth. The parameters $A$ and $\Delta_{j}$ must be chosen to ensure the continuity between the two portions of the curve. It is well accepted that the heavy hole temperature is close to the lattice one under moderate to high electric fields $[53]$. An excellent reproduction of the photoluminescence spectra recorded under applied voltage is obtained using Eqs. (1–2), considering the main $j \rightarrow k$ transitions with the largest overlap integral and leaving $E_{p}$, $T_{e}^{j}$ and $A_{jk}$ as fitting parameters. A detailed analysis of THz QCLs based on different active region designs has then been performed:
Figure 4. Differences between the mean lattice temperature $T_L$ and the heat sink temperature $T_H$ (●) and between the electronic temperatures in the active region subbands $T_e^j; j = 1, 2$ (○), 5 (upper lasing state ▲) and $T_H$ for a RP THz QCL measured as a function of the electrical power [44]; (b) Differences between the mean lattice temperature $T_L$ and the heat sink temperature $T_H$ (●) and between the miniband electronic temperatures $T_e^{M1,M2}$ and $T_H$ (●) in the active region of a BTC THz QCL measured as a function of $P$ [51]. (c) Differences between the mean lattice temperature $T_L$ and the heat sink temperature $T_H$ (●) and between the electronic temperatures of the injector states $T_e^{j}; j = inj$ (●), and of the active region $j = act.$ (▲) and $T_H$ in a THz QCL with a hybrid active region scheme measured as a function of $P$. The lines are linear fits to the data. The shaded areas mark the lasing region [55].

- **RP THz QCLs:** the electronic temperatures of all subbands, excluding the upper state, are nearly equal to the lattice temperature and increase linearly with the dissipated electrical power ($P$) with a slope of tens of K/W, slightly larger than the thermal resistance. On the other hand, the temperature of the upper laser level can reach 100 K more than the lattice one (Fig. 4a). In total, 25–40% differences in the subband electronic temperatures have indeed been predicted by Monte–Carlo simulations [54]. This effect is ascribed to the reduced efficiency of inter-subband e–e scattering channels coupling electrons in the upper state and in the injector states, with respect to intra-subband e–e processes. One important implication of this finding is that the high upper radiative state temperature leads to relatively fast non-radiative relaxation times $\tau_{2 \rightarrow 1, j} = 1$ ps (see Figure 1c), representing a key limiting factor for the high temperature operation of RP devices.

- **BTC THz QCLs:** the electronic temperatures of subbands belonging to the lower ($M_1$) and higher ($M_2$) active region minibands are nearly equal and increase linearly with $P$ with a slope larger than the thermal resistance ($R = dT_e/dP = 20.1$ K/W in the data of Fig. 4b). Hence, the vast majority of electrons share here the same electronic temperature. Due to the excitonic nature of the PL peaks arising from electrons that populate the upper laser level $b$, the electronic temperature of this level cannot be extracted precisely [51,52].

- **Hybrid THz RP-BTC QCL design:** at very low $P$ values, i.e. when the doublet of lowest injector states is close to resonance with one of the
four subbands in the extraction miniband, the energy redistribution process between the injector and the miniband is so efficient (e-e scattering time $\tau_{ee} \sim 100$ fs) that a common temperature between the injector and active region subbands is reached. At higher $P$ values electrons are progressively injected into the upper lasing state and a common electronic temperature is set among different active region subbands, which increases with $P$ at a much faster rate than in region I. Cold electrons progressively populate the upper state and are scattered elastically or quasi-elastically with a large excess energy to a lower state in the active region, therefore thermalizing within their respective subbands at a temperature $T_e^{\text{active}} > T_e^{\text{inj}}$. This is a consequence of the fact that the carrier density in the active region is always one order of magnitude smaller than in the injector states. A large negative discontinuity in the differential resistance at the onset of region III indicates that the laser threshold is reached while the population inversion $\Delta n$ becomes clamped at the onset for stimulated emission [55,56]. Correspondingly, the electron heating rate decreases (region III) meaning that the photon emission extracts part of the input power, efficiently cooling the electrons. A further proof of the optical origin of the latter effect is the fact that the change in the electronic temperature slope disappears when the same measurements are performed on a non-lasing mesa device [55]. In region IV lasing ceases, as revealed by the sudden jump in the differential resistance (Fig. 4 c) [55].

- **SA THz QCL design**: the injection here takes place into an ancillary state at energy higher than the upper laser level by approximately one LO phonon energy [14], leading to a very different electron temperature dynamics. While in the conventional RP structures electrons are directly injected into the upper laser level, causing a large extra heating with respect to the lattice ($\Delta T \sim 70–110$ K) or the ground level ($\Delta T \sim 100$ K) [44], in the SA scheme both laser levels remain much colder (by a factor of 3–5) and share the same electronic temperature of the ground level ($j = 1$). The injector state designed at a larger energy separation is on the contrary $\sim 45–60$ K hotter than other levels at large dissipated powers. In addition, the radiative subbands population ratio shows an efficient lower laser level depletion at low electronic temperatures, whereas at temperatures larger than $T_e \sim 180$ K the ratio $n_2/n_1$ starts to increase with a rate comparable with the thermal activation one [57].

5. Temperature performance

Nearly two decades after the first demonstration of THz QCLs, room temperature operation still remains a top priority. Several theoretical
models have been employed to understand the details of charge transport and optical gain within THz QCLs, based on various approaches such as rate equations [58], density matrix (DM) [59–61], non-equilibrium Green function [62,63], and Monte Carlo (MC) techniques [64–67]. Via a careful optimization of gain, oscillator strength and population inversion as well as a careful reduction of waveguide losses, operation has slowly inched up to 200 K [68] in pulsed mode and 129 K [69] in continuous wave, already several years ago. Thanks to the recent development of water-cooled five-stage thermoelectric refrigerators, this performance has already proven sufficient to operate THz QCLs without any cryogenic equipment, achieving respectable peak output powers of a few mW at duty-cycles of 2–5% [70].

From a physical viewpoint, there are various processes that are expected to cause the degradation of population inversion (and gain) in THz QCLs at higher temperatures. Backfilling of the lower laser state is mainly affected by thermal excitation from highly populated levels or by hot-phonon effects [22,71]. Another path is due to the onset of thermally activated LO-phonon scattering, as electrons in the upper subband acquire sufficient in-plane kinetic energy to emit a LO-phonon and relax to the lower subband [72–75]. Both of these mechanisms sensitively depend on the electronic temperature that may significantly exceed the lattice one [43,50,51]. Further detrimental processes include temperature broadening [76–78] and thermal leakage of carriers into continuum states [79].

In order to properly take into account all these effects, an advanced modelling scheme based on non-equilibrium Green’s functions [80,81] has been employed already at the active region design stage to maximize the achievable gain at high lattice temperatures up to 300 K [17]. Focusing on the simple but advantageous two-well concept discussed above, which had recently shown close to record operating temperatures [82], the optimization of doping and layer thicknesses [17], together with the adoption of a low-loss Cu-Cu waveguide and a systematic study of the influence of the geometrical cavity parameters and wafer uniformity, has yielded devices lasing above 210 K with peak powers still in the mW range on a relatively conventional 4-stage Peltier cooler (see Figure 5) [83]. This result finally advanced the record temperature after seven years and offers interesting perspectives for further progress; only a subset of the actual device parameter space has, in fact, been explored and other aspects (barrier composition, doping position, waveguide fabrication, etc.) could be optimized and lead to higher temperature lasing. We want to remark that for these THz QCLs the lattice thermal energy is already greater than the photon energy \( k_B T > h\nu \), thereby confirming that no limitation is to be expected from a device operating far from equilibrium conditions.
In the course of writing this manuscript, we became aware that the MIT group has achieved a new record high operating temperature of 250 K in a similarly designed THz QCL [84].

Alternatively, room temperature emission at THz frequencies from a QCL can be achieved via intracavity difference-frequency generation (DFG) from mid-infrared (mid-IR) QCLs. This is currently the only electrically pumped semiconductor source capable to cover the entire 1–5 THz range at room temperature, in continuous-wave, with power outputs of 14 μW [85].

6. Distributed feedback lasers

The availability of stable, single-mode emission, at a precise frequency, is crucial for many applications and is thus an important prerequisite for the use of lasers in areas like high precision spectroscopy, chemical sensing and astronomy. A feedback at a precisely defined frequency can be provided by periodically structuring the laser cavity: the well-defined frequency originates from the scattering of light at many locations along the cavity so that feedback is created by constructive interference. Because the feedback is, in this case, distributed along the entire laser, such devices are termed distributed feedback (DFB) lasers [86].

An easy way to understand a DFB resonator is to consider the situation in k-space (Figure 6). A periodic structure leads to a well-defined Bragg peak in the grating Fourier spectrum, off which the photons are backscattered if the condition \( k_p = n k_B - k_p \) is fulfilled, where \( n \) is called the order of the grating, and \( k_B \) and \( k_p = n_{\text{eff}} \omega / c \) are the Bragg peak wavevector and that of the free waveguide photon, respectively. Note that feedback can also be produced through first-order scattering off a Bragg peak of higher order, which are

![Figure 5](image-url). Left: light- and voltage current characteristics of the high temperature THz QCL of Ref. 83. Right: threshold current density as function of temperature and picture of the Peltier-cooled laser head (inset). Reproduced from [83] with the permission of AIP Publishing.
always present in non-sinusoidal grating profiles. The coupling constant $\kappa$ that quantifies the coupling between the forward and backward traveling mode of the waveguide is defined as:

$$\kappa = \frac{\pi \Delta n_{\text{eff}}}{4 n_{\text{eff}} \Lambda} + i \frac{\Delta \alpha_w + g_{\text{th}} \Delta \Gamma}{2} \quad (3)$$

where $\Lambda$ is the grating period and $g_{\text{th}}$ is the threshold gain [86]. The periodic modulation of refractive index along the waveguide is assumed to be of the form $n_{\text{eff}}(z) = n_{\text{eff}} + \Delta n_{\text{eff}} \cos(z)$, with a similar behavior for waveguide losses and confinement, whose modulation amplitudes are respectively $\Delta \alpha_w$ and $\Delta \Gamma$. The coupling constant $\kappa$ represents basically the exponential decay constant of the propagating wave in the grating, and, therefore, single-mode operation is typically obtained when $|\kappa| L \approx 1$, where $L$ is the length of the laser. The laser is called index-coupled if the coupling constant is mostly real, loss-coupled if it is imaginary, or complex-coupled if both parts contribute.

The integration of distributed feedback resonators within THz QCLs is simple because the optical mode peaks at the metallic cladding on top of the active region, and, therefore, is strongly influenced by any modification of this boundary layer. The top metallization, in turn, can easily be modified by optical contact lithography, especially because the typical DFB-period is in the tens of microns. DFB resonators developed so far for THz QCLs are based on various scattering mechanisms, like the periodic incorporation of annealed-contact-, photoresist-, or nickel-based stripes across the waveguide [87–90]. These perturbations, which leave the top-metal of the

Figure 6. Photonic bands in a DFB laser. Depending on which bandgap overlaps with the gain spectrum, one speaks of first, second, or third-order DFB. In the second-order case the in-plane wavevector $k$ is zero and the resulting emission is vertical. In the third-order case, $k$ is exactly at the edge of the light cone (and hence gives rise to purely in-plane emission) if $n_{\text{eff}} = 3$. 

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waveguide uninterrupted, are especially suited to single-metal resonators, since they provide sufficient coupling between forward and backward traveling wave, but do not heavily affect the overall losses or confinement factor.

Stronger coupling mechanisms can be designed by directly patterning the metal, showing single-mode emission in gratings with less than 100 periods. The slit width can be used to design the coupling strength, and it was found that grating duty cycles of around 90% give a strong coupling constant without affecting the performance too much [91]. By far the strongest variation in effective refractive index is actually obtained if, beyond slotting the top metallization, also the top-contact layer or even the whole semiconductor active material of the laser is etched. With this fabrication, stable single-mode emission from gratings with only a few tens of periods has been achieved.

In general, DM THz QCLs with slits in the top metallization are prone to lasing on a different transverse mode, owing to the higher losses introduced to the central part of the waveguide by the grating. The first DFB structure therefore was here realized with a laterally corrugated waveguide, controlling precisely also the grating position with respect to the facets [92]. The very strong influence of the facets, in fact, makes it very hard to transfer the simple grating designs from single- to double-metal waveguides. Furthermore, for DM waveguides, simple DFB models like the coupled-wave theory or the transfer-matrix formalism are not very helpful, because the individual slit scattering mechanism and the very strong perturbation by the facets are difficult to be quantitatively incorporated. Furthermore, potential surface losses cannot be inferred from these simple models. A finite element numerical method, in contrast, directly solving the Helmholtz equation, treats a much wider class of problems, and is then often the method of choice for double-metal DFB lasers [93].

Very good results have been achieved with the use of so-called third-order DFB gratings [35]; lasers based on this concept, in fact, combine nice single-mode operation with highly efficient collimated emission. The idea is to have the device lasing at a photonic band-gap at the edge of the grating 1D Brillouin zone for the feedback, and at an energy matching exactly the air/vacuum light-line to ensure perfect extraction from the whole laser length (see Figure 6). Clearly, given the larger than one effective waveguide refractive index $n_{eff}$, both requirements can be met only from the third bandgap on and if $n_{eff}$ is equal to the order of the bandgap. This can be obtained for instance by precisely tailoring the size and depth of the DFB slits so as to tune $n_{eff}$ [35].

The concept has been extremely successful, allowing so-called photonic wire lasers with lateral dimensions much smaller than the wavelength [94], and, consequently, very low drive current. It has subsequently been
optimized using the analogy to microwave transmission lines with integrated antennas, yielding high slope efficiencies of the order of 0.5 W/A [95,96] and implementing asymmetric DFB unit cells to ensure unidirectional emission [97]. Alternatively, two different gratings can be implemented in the same laser cavity, a conventional first-order one to provide the feedback and a second one supplying the necessary wavevector to scatter the light out from the surface at a chosen angle, in either a standard [98] or a corrugated cavity configuration [99], overcoming the issue of precise refractive index matching of 3rd order DFBs (see Fig. 7).

Recent research in THz DFB lasers has intertwined with that aiming at realizing vertically emitting devices [93]. Starting from the second-order gratings already mentioned above [32–34], further, more complex DFB structures have been conceived, like photonic heterostructures [100] or arrays of phase-locked DFB elements [101,102]. A further novel approach relies on the development of vertical-external-cavity surface-emitting THz QCLs (VECSELs) comprising an amplifying metasurface reflector composed of a sub-wavelength array of antenna-coupled quantum-cascade sub-cavities, that can operate on their first-order (TM01) resonance [103],

![Figure 7](Image).

**Figure 7.** Device pictures and far field emission profiles of low divergence THz wire DFB lasers. a) 3rd order DFB (reprinted by permission from Springer Nature: [35] copyright 2009); b) corrugated wire laser [99].
or on their third-order lateral modal resonance (TM03), allowing, in this latter case, to reach THz output powers up to 1.35 W [104].

7. Quasi-crystal and random lasers

Photonic engineering, combined with new resonator concepts, has enabled the performance of THz QCLs to be designed with an incredible level of control, offering a flexible platform to tailor the emission spectrum, the beam profile and the output power, simultaneously. As a typical example, quasi-crystal THz lasers, loosening the distinction between symmetric and anti-symmetric modes allow circumventing the typical photonic crystals issue of power cancellation in the far-field, achieving a remarkably high power extraction (240 mW), ≈720 mW/A slope efficiency and good beam collimation (<10° divergence), both in single-mode and multimode regimes [40,41,105]. Very recently, random THz lasers (RLs) have been also successfully developed [106–110], in either 2D or 1D [109] geometries. They strongly differ from conventional lasers, which traditionally comprise a gain medium enclosed in an optical cavity to produce feedback. Although RLs also require an active medium, this no longer needs to be embedded in a specially designed cavity, since the feedback mechanism arises from multiple elastic scatterings in the highly disordered structure [111]. In RLs, the emitted photons can be amplified and scattered many times in the gain medium, resulting in a rich interference scheme, with each optical mode having a different degree of localization. This complex interplay between the intrinsic disorder and the nonlinearity of a random active medium [111,112] also gives rise to many interesting physical phenomena, such as gain competition and nonlinear wave mixing of the optical modes [113].

THz QCL random lasers have been demonstrated by employing different device geometries exploiting air pillars [106], semiconductor pillars [107] or a combination of semiconductor and metal pillars [108], all etched through the entire active region, or through a disordered sequence of scatterers [109] only implemented in the upper metal and highly doped semiconductor cladding, leaving the active region core unperturbed, and enabling multimode emission in continuous wave over a 450 GHz bandwidth. Figure 8 displays pictures of THz quasi crystal or random lasers.

8. Frequency tuning

A relevant progress direction has been targeting the external tuning of the single-mode emission. Thermal and electrical tunability through joule heating in THz QCLs, in fact, remain relatively low (generally of the order of just few GHz), with typical tuning rates of several tens of MHz/K or of a few MHz/mA [114]. Among the two, in general electrical tuning is preferred,
since it allows much higher scanning speed and improved stability, often crucial aspects in spectroscopic applications.

An optomechanical tuning concept demonstrated by Q. Qin and co-workers [115] showed a single-mode continuous tuning broad enough to cover the entire laser gain spectrum. This approach is based on the fabrication of first-order single-mode DFB lasers in a narrow strip double-metal geometry, having transverse dimensions $w$ much smaller than the wavelength ($w = \lambda/3$), with a strong sinusoidal first-order Bragg grating etched into one side of the ridge. The DFB geometry then fixes the longitudinal wave vector of a specific resonant mode in the waveguide. While the single lasing mode is selected from the narrow ridge and the design of the grating, the tuning is obtained by changing the transverse wavevector $k_\perp$ by mechanically bringing another material (plunger) close to the flat side of the laser ridge, thus influencing the mode index (Fig. 9a).

More recently, the same group developed a new solution combining the fabrication of still narrower lasers ($\lambda/8$) with the use of plungers based on micro electro-mechanical systems (MEMS) [117]. With this elegant technical approach, a continuous and reversible tuning range of 330 GHz in single-mode has been obtained [117].

In a similar concept, the DFB mode can be coupled to an external microcavity through a top 2$^{nd}$ order grating and laser emission can then be tuned by changing its size by using a movable mirror [118,119]. With this technique 162 GHz single-mode continuous tuning from a surface-emitting THz QCL has been demonstrated [119]. Alternatively, by exploiting a coupled-cavity architecture with a precisely positioned defect lattice that engineers the free spectral range and finesse of the cavity, and by employing Vernier selection rules, frequency tuning over 209 GHz has been demonstrated, including mode hop-free continuous tuning of $\sim 6 - 21$ GHz across

Figure 8. Device pictures of disordered THz QCL resonators with a quasi-crystal structures implemented in a) 1D [105] or b) 2D [40] cavity geometry and c) picture of a random THz laser [110].
six frequency bands, controlled through Stark shift, cavity-pulling, localized Joule heating, or thermal effects [120].

Very recently, THz quantum cascade metasurface-based VECSELs exhibiting 20% continuous fractional tuning of a single laser mode have been demonstrated. The tuning is here allowed by the subwavelength thickness active metasurface, which induces lasing on very low-order Fabry–Pérot cavity modes. Good beam quality and high output power have been simultaneously obtained (Fig. 9b-d) [116].

All these electro-mechanical approaches are also compatible with moderately high tuning speeds, although they clearly lack the simplicity of a direct current modulation.

9. THz quantum cascade laser frequency combs

Broadband QCLs, with heterogeneous [121–123] or homogeneous [124–126] active region designs, have been recently demonstrated to operate as stable chip-scale THz optical frequency comb synthesizers (FCs), characterized, in the frequency domain, by a set of equidistant spectral lines, which share a well-defined and stable phase relationship between one another.

The large third-order $\chi^{(3)}$ Kerr nonlinearity of the active medium indeed gives rise to the interaction between adjacent modes via four wave mixing (FWM) resulting in a frequency modulated FC (see Fig. 10). However, in a free-running THz QCL, the modes are not uniformly spaced, owing to chromatic dispersion. As a result of a frequency-dependent refractive index, the laser free spectral range is index dependent, producing an unevenly spaced spectrum.
The interplay between FWM and group velocity dispersion (GVD) in the laser cavity plays a major role in determining the stability of the QCL FC over the laser operation range. Gain medium engineering can indeed allow a flat top gain, however only at a specific bias point, meaning that the nature itself of that gain medium entangles the dispersion dynamics at other biases. As a result, FC operation can occur over a small fraction of the operational current, usually less than 20% of the lasing range [121,122,124,127].

Handling such bias-dependent dispersion compensation at THz frequencies is a very challenging task. Present attempts include either passive or active schemes. In the first case, the laser bar can be shaped with an integrated dispersion compensator [124] and allows achieving FC operation over only 29% of the QCL operational regime. Alternative configurations include tunable Gires–Tournois interferometers (GTI), comprising a movable gold mirror back coupled with the QCL, which leads to a further 15% increase of the regime where the THz QCL operates as an FC [128], as a result of a partial compensation of the total group delay dispersion (GDD).

Active approaches rely on the use of external cavities, as coupled dc-biased sections [129] or Gires–Tournois interferometers [130], allowing either a full coverage of the bias range, at the price of a reduced optical bandwidth (≤400 GHz) and negligible (µW) optical power outputs [129], or only a specific operational point [130].

A preliminary assessment of the FC operation in a THz QCL can be done through a characterization of the bias-dependent intermodal beat-note (IBN). The presence of a single and sharp (linewidths in the kHz range) IBN represents a good precursor for a genuine comb operation. However, such a technique lacks information about the stability of the Fourier phases, that can be extracted via alternative optical techniques as the shifted-wave
interference Fourier-transform spectroscopy (SWIFTS) [131,132], which gives access to the phase domain by measuring the phase difference between adjacent FC modes or via the Fourier analysis of the comb emission (FACE) [123,133]. Relying on a multi-heterodyne detection scheme, this technique is capable of real-time tracing the phases of the modes emitted by the FC, thus working even if the comb presents spectral gaps.

The research field of THz frequency combs is extremely vibrant and promises major impacts in several application domains crossing quantum metrology, dual-comb spectroscopy [134], hyper spectral imaging, time-domain nano-imaging, quantum science and technology, and non-linear optics [135].

10. Intrinsic linewidth of THz QCLs

In a QCL, the laser linewidth (LW) can be expressed by a slightly modified version of the Schawlow–Townes (ST) formula [136], including the linewidth enhancement factor ($\alpha_e$), which takes into account the refractive index variations with gain, caused by electron density fluctuations [137]:

$$\delta v = \frac{1}{4\pi} \frac{(1 + \alpha_e^2)^{\gamma^2} \alpha_m(h\omega)}{2P_{out}\alpha} n_{sp}$$  \hspace{1cm} (4)

where $\gamma = \nu_g\alpha$ is the cold cavity linewidth, $\alpha$ accounts for the total cavity losses, $\alpha_m$ represents the mirror losses, $\nu_g = c/n_{\text{eff}}$ the group velocity, $c$ the speed of light in vacuum, $n_{\text{eff}}$ the effective refractive index, $n_{sp}$ the population inversion. A theoretical model, recently developed for QCLs [138], demonstrates that the latter formula can be further refined including the contribution of thermal (black-body) radiation, playing a key role in the far-infrared. In a low electron temperature regime (<100 K) [51,52] and once the operating pumping conditions are below a specific limit, set by the relaxation dynamics between electron and lattice bath, the THz QCL LW can be rewritten as:

$$\delta v = \frac{1}{4\pi} \frac{(1 + \alpha_e^2)^{\gamma^2} (h\omega)}{P_{\text{int}}} (1 + 2N_{bb}) = \frac{1}{4\pi} \frac{(1 + \alpha_e^2)^{\gamma^2} \frac{\beta \tau_f}{\tau_s}}{(1 - \frac{1}{I_{\text{th}}})} (1 + 2N_{bb})$$  \hspace{1cm} (5)

where $n_{sp}$ has been assumed equal to 1, $P_{\text{int}} = \alpha/\alpha_m$ $2P_{out}$ represents the photon number in the laser cavity (or internal power), $N_{bb}$ is the thermal photon population, ($\beta/\tau_s$) the spontaneous emission rate coupled into the lasing mode and ($1/\tau_s$) the total relaxation rate of the upper level. Regardless of the specific gain medium architecture, Eq. 5 allows to calculate the QCL LW once the device internal efficiency or the gain media/operating condition parameters are known.
Recently, the spectral purity of a THz QCL has been investigated via the measurement of its frequency-noise power spectral density (FNPSD), providing an experimental evaluation and a theoretical assessment of its intrinsic LW. Intensity measurements were performed to retrieve information in the frequency domain by converting the laser frequency fluctuations into detectable intensity (amplitude) variations [139]. As a discriminator, the side of a Doppler-broadened methanol molecular transition centered at $v_o = 2.5227816$ THz was used. Given the intrinsic low-noise nature of the measurement, the converter (or discriminator) must introduce a negligible noise providing, at the same time, a gain factor suitable for a good detection. A schematic diagram of the experimental set-up is shown in Figure 11.

The full spectrum of the QCL was obtained by sticking together several acquisitions taken in smaller spectral windows, in order to ensure a high overall resolution. The resulting FNPSD spectrum is plotted in Figure 11, together with the current-noise power spectral density (CNPSD) of the current driver, converted to the same units by using the current tuning coefficient. The asymptotic flattening of the FNPSD above 8 MHz, with its visible deviation from CNPSD, indicates a flattening to a white noise level ($N_w$), leading to a FWHM $\delta v = \pi N_w$ from which an intrinsic LW $\delta v = 90 \pm 30$ Hz [138] has been extracted. Despite the broadening induced by thermal photons, the measured linewidth results narrower than that found in other semiconductor laser, to date.

Controlling the linewidth of THz QCLs and pushing it down to its quantum limited value can pave the way to exciting new research. For

![Figure 11](image_url)

**Figure 11.** *Left:* Schematic diagram of the experimental setup, where the function of the molecular line acting as frequency-to-amplitude discriminator is shown. *Right:* Experimental FNPSD of the THz QCL (orange trace), compared to the contribution to frequency-noise of the CNPSD of the current driver (blue trace). The dashed line marks the white noise level. From Ref. 139.
example, rotational transitions in molecules can lead to natural LW of a few hundred hertz. Comparably narrow lasers are therefore required to interrogate or manipulate cold samples to obtain new insights, for example into the measurement of fundamental constants by means of highly accurate frequency measurements [140].

**Conclusions and perspectives**

Almost 20 years after their first demonstration, THz QCLs have become a mature technology that is on the verge of widespread commercialization. The spectacular progress in operating temperature achieved in the last year will certainly represent the final driving achievement in this direction, freeing QCL-based THz systems from the necessity of costly cryogenic equipment and opening up new applications where portability is an important property.

Yet, THz QCLs still remain a very intriguing research field even from a more scientific point of view. The possibilities offered by QCL frequency combs and the physics underlying their operation are just beginning to be tapped and will certainly enable new investigations in a variety of fields from metrology to sensing. New classes of materials are now getting closer to impact THz QCL development: on one side Ge/SiGe structures are showing more and more promising intersubband response and transport properties, making them potential candidates for the next generation of THz QCL active regions; on the other hand, 2D materials like graphene, with the extreme plasmonic confinement it can provide, may soon become a new viable solution for ultra-small THz QCL resonators. These are just two examples of innovative elements that are likely to tremendously enrich the physics of the THz QCL in the near future, opening new avenues of research for this already very successful device.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

**Funding**

The work was partially supported by the European Research Council through the ERC consolidator grant SPRINT (681379). This research was sponsored in part by the NATO Science for Peace and Security Programme under grant G5721 THESEUs.

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