

Quantum cascade lasers: 20 years of challenges

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Abstract: We review the most recent technological and application advances of quantum cascade lasers, underlining the present milestones and future directions from the Mid-infrared to the Terahertz spectral range. Challenges and developments, which are the subject of the contributions to this focus issue, are also introduced.

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1. Introduction

The quantum cascade laser (QCL) is an unipolar device that exploits optical transitions between electronic states (conduction subbands) created by spatial confinement in semiconductor multi-quantum-wells, via the quantum engineering of electronic wavefunctions on a nanometer scale. The extreme precision of the material growth necessary to obtain the particular characteristics required by the device design, together with the large number of layers and the complexity of the structure, makes this laser perhaps the most impressive demonstration of bandgap engineering.

Conceived and demonstrated at Bell Laboratories in 1994 [1], QCLs rapidly evolved across 20-years of discoveries from an interesting laboratory proof-of-principle to a powerful and groundbreaking technology having concrete impacts in many technological applications such as trace gas analysis, optical communications and real-time imaging.

The history of QCLs is full of exciting milestones. The use of "inter-subband transitions" for radiation amplification, the core idea of the QCL, was first proposed in 1971 by *Kazarinov* and *Suris* in a superlattice structure [2]. It was the later development of growth techniques such as Molecular Beam Epitaxy (MBE) [3] or metallorganic vapor phase epitaxy (MOCVD) [4] that provide unprecedented control of layer thickness down to a single atomic monolayer that has enabled realization of this concept. In essence, new materials are created by the use of heterostructures which creates sharp discontinuities in the conduction and valence bands edges [5]. This signified the birth of a new class of phenomena and devices, offering novel ability to control the size quantization effects on charge carriers at nanometer scale and

leading to the tuning of the electronic energy by variation of the layer thickness and control of the electron localization and transport as well as of the optical processes.

The most characteristic feature of the QCL is the possibility to tune the emission frequency over a large bandwidth without changing the semiconductor material system, merely by, changing the size of quantum wells to change the energy separation of electronic states. This, together with the unipolar nature of the charge transport and the peculiar shape of the density of states, is in stark contrast with conventional inter-band lasers, where the emission wavelength depends on the material bandgap and the gain is strongly temperature-dependent. In addition, the multistage-cascaded geometry allows for electron recycling, so that each electron injected above threshold may generate a number of photons equal to the number of stages.

It has been well established both theoretically and experimentally [6,7] that in a QCL electron- longitudinal optical (LO) phonon scattering is the dominant inter-subband scattering mechanism for subband separations wider than the LO-phonon energy E_{LO} . For intrawell transitions, these scattering times tend to be less than 1ps. For inter-subband transitions where the separation is less than E_{LO} , emission of LO-phonons is energetically forbidden at low temperatures. Non-radiative relaxation is therefore dominated by the combination of electron-electron ($e-e$) scattering, electron-impurity scattering, and 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 [8–10]. Furthermore, the electron scattering with ionized impurities plays an important role in intersubband transport, and in some cases may be stronger than $e-e$ scattering. Other relaxation paths turn out to be less important: acoustic phonon scattering is relatively inefficient (100 ps scattering times), especially at low-temperatures where the phonon population is small [11]. Interface roughness scattering competes with optical phonons in the scattering efficiency, and becomes extremely efficient for short wavelength transitions above 300 meV. Interface roughness through intra-subband scattering has also found to be the main broadening mechanism for optical transitions between intersubband states, whose width is generally not lifetime limited [12]. Other fundamental channels to be considered are represented by intrasubband - type transitions. Intrasubband LO-phonon scattering is an important process in cooling the subband electron gas [9]. The effect of intrasubband $e-e$ scattering is to thermalize the electron distribution inside a particular subband.

In principle, QCLs may be realized using almost any semiconductor material system. To date, the best QCL performance has been obtained by using four semiconductor material systems: GaInAs/AlInAs grown on InP substrates [13], GaAs/AlGaAs grown on GaAs substrates [9], AlSb/InAs grown on InAs [14,15], InGaAs/AlInAsSb, InGaAs/GaAsSb or InGaAs/AlInGaAs grown on InP substrates [16–19]. The intersubband gain affected by the material choice since it is influenced by both electron effective masses m^* in well and barriers [20]; the conduction band ΔE_C limits the shortest possible wavelength operation for a given material system

After two decades from their invention, QCLs operating in the mid-IR have reached impressive performance levels. Multi-watt output power, continuous wave (CW), room-temperature (RT) devices operating across the mid-infrared (IR) with wall-plug efficiencies of up to 21% observed at room temperature, and larger than 50% cryogenic temperatures (77 K) [21–23]. Spectral coverage has been achieved at wavelengths from ~ 3 –25 μm with the potential for large tunability [24]. QCLs with new promising material systems have been recently demonstrated to work up to 400 K at wavelengths within the first atmospheric window (3–5 μm) [15,17,24–26]. In 2002, the spectral coverage of QCLs was extended to the Terahertz (THz) region [27], where efficient and miniaturized sources operating in the 1.2–4.9 THz window have now been successfully developed, in either single-plasmon or double-metal waveguide configuration [28].

The present state of available frequencies as a function of the temperature performances of QCLs is schematically summarized in Fig. 1.

In this manuscript we will review such recent advances in the field of mid-IR and far-IR QCLs, from the major technological developments to some of the present challenging applications.

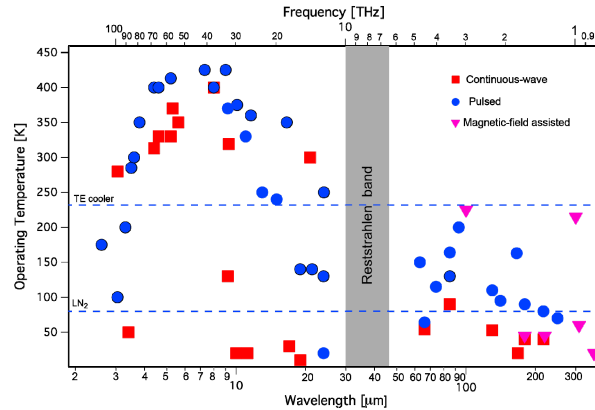


Fig. 1. Operating temperature plot as a function of the emission wavelength (or frequency, top axis) for quantum cascade lasers.

2. Mid-IR quantum cascade lasers

In the past few years, Mid-IR QCL research has progressively shifted from the lab to the photonic market: many commercial providers now offer QCL and interband cascade lasers (ICLs) in different configurations ranging from Fabry-Pérot devices, to distributed feedback (DFB) resonators, to multi-wavelength systems based on tunable external cavities, as well as high-power devices. Remote sensing [29], metrology [30] and infrared countermeasures are some of the most exciting areas where QCL technology finds progressively more and more space, resulting an enabling platform. Here we will discuss some very recent advances in the field, namely high performance Mid-IR QCLs, the realization of on-chip frequency combs in the Mid-IR based on QCLs and the photonic engineering to address tunability and integrated solutions for spectroscopy and sensing applications.

2.1 High performance Mid-IR quantum cascade lasers

Room temperature, CW operation of Mid-IR QCLs has been achieved more than 10 years ago [31]. The last decade saw a dramatic improvement of the QCL performances across the Mid-IR range. The development of high performance Mid-IR QCLs in the last few years [23,31] has made possible to reach remarkable performances with emitted powers in the 1-5 W range. The wall-plug efficiency, namely the ratio between injected power and extracted optical power in a laser device, has reached values as high as 27% in pulsed mode and 21% in CW mode [23] with more than 5 W of CW output power at RT employing advanced active region engineering in the InGaAs/AlnGAs/InP material system. In several high performance designs the conduction band offset has been engineered in order to limit the leakage current above the barriers and to enhance the upper state lifetime through a careful choice of barrier and well heights [33,34]. These impressive performances are then due to a combination of refined quantum engineering of the laser active region together with a refined material growth, advanced processing solutions like buried heterostructures and careful management of dissipated heat. Such remarkable power performance has already allowed the development of RT, Mid-IR-based THz sources relying on intra-cavity difference frequency generation [35,36] In the short-wavelength Mid-IR region the most relevant results come from advanced engineering of materials and from ICL devices. The conduction band offset limits the shortest

possible wavelength achievable in a particular material system – as the upper radiative subband energy approaches the barrier height, increasing numbers of electrons are lost to thermionic leakage. Hence, QC-lasers with photon energies above 250 meV ($\lambda < 5 \mu\text{m}$) have required using of the InGaAs/AlInAs/InP alloys adopting strain balanced structures [13] or introducing AlAs barriers to increase the confinement. The use of different antimony-based ternary alloys [37] has led to operation above room temperature at wavelengths around 3 μm and to the demonstration of laser emission below 2.7 μm [38] using InAs/AlSb heterostructures.

While not technically a QCL, the interband cascade laser (ICL) is a competitive approach for emission wavelengths from approximately 2–4 μm . First demonstrated in 1995 [39], the ICL is based on band-to-band transitions in heterostructures exploiting type-II band alignment of the GaSb-based material system. ICLs now display excellent performances reaching 15% of wall plug efficiency in CW at room temperature with 290 mW power at 3.6 μm [40]. On the other extreme of the Mid-IR spectrum, close to the phonon absorption band the same material combination of Sb-based heterostructures has been successfully employed to improve the performance of long-wavelength Mid-IR lasers operating at a wavelength of 17 μm or longer [41]. Recently, QCL emission at 24.4 μm and at 240 K has been demonstrated by using InGaAs/AlInAs material system [42].

2.2 Frequency comb operation of quantum cascade lasers

Optical frequency combs [43] have revolutionized the fields of high precision spectroscopy and metrology and their paramount importance has been certified by the attribution of the 2005 Nobel prize in physics to Ted Hänsch and John Hall. The extension of the comb concept to all the electromagnetic spectrum is currently involving a number of technologies, including quantum cascade lasers. Mid-IR frequency combs have been already realized starting from pulsed sources by means of non-linear optics and frequency down-conversion. QCLs are an ideal platform to implement a fully on-chip, comb-based Mid-IR spectrometer. In the last three years a number of milestones have been achieved, starting with the first demonstration of comb operation of a broadband mid-IR QCL [44]. Like in micro-resonator Kerr combs [45], the comb-driving mechanism in QCLs is the large intrinsic $\chi^{(3)}$ non-linearity of the gain medium which favours phase locking due to cascaded four-wave-mixing processes. Therefore, Mid-IR QCL combs are practically CW sources, differently from the combs produced by conventional mode-locked lasers where broadband, ultra short optical pulses are generated. Group velocity dispersion can be significantly reduced through active region design and waveguide design. After the first demonstration, theoretical analysis has been carried on to identify the QCL comb formation mechanism [46] and dual-comb spectroscopy demonstrated [47]. QCL comb operation has been recently extended also to the THz range, where the role of dispersion is much stronger than in Mid-IR: the careful compensation of this effect has led to QCL combs at 3.5 THz [48].

2.3 Photonic engineered and integrated solutions for high tunability and lab-on-a-chip.

The area of QCL-based integrated devices for spectroscopic applications is another research area that has shown significant recent progress. Arrays of vertical-emitting ring cavities [49] and DFBs in MOPA configuration [50] represent promising solutions for highly integrated spectrometers and sensing systems. Interestingly, the intersubband transitions can be successfully used to produce detectors matching the spectral range of the quantum cascade lasers. Quantum Well Infrared Photodetectors (QWIPs) [51] and Quantum Cascade Detectors (QCDs) display very good performances in the Mid-IR [52]: they start to be combined with photonic microstructures to increase the responsivity and can be integrated together with QCLs to form an all-intersubband high performance sensing platform [53,54]. Due to the atomic-like joint density of states characteristic of intersubband transitions, QCLs are particularly suited for broadband operation since the gain profile can be engineered without incurring in reabsorption. Heterogeneous QCLs [55] are the core of broadly tunable external

cavity laser systems [56], which nowadays represent a sizeable portion of the QCL products on the market. Recently, tuning ranges of more than a 1000 cm^{-1} from a combined source (Block Engineering) and more than 350 cm^{-1} from a single chip (Daylight solutions) have been achieved and made commercially available.

3. Terahertz quantum cascade lasers

The QCL provides a very attractive approach to cover the upper terahertz frequency range – at this time THz QCLs of various types have now been demonstrated to cover the wavelength range from 1.2 to 4.9 THz. Power levels are typically on the level of a few milliwatts, although large area devices have recently demonstrated Watt-level output powers [57,58]. THz QCLs are qualitatively different from the mid-infrared ones for two reasons. First, the photon energy is smaller than the LO-phonon energy, which leads to significant differences in carrier dynamics, and requires a more precise injection and depopulation of carriers into the closely spaced energy levels. Hence, a different set of active region designs are used. Second, because free carrier loss scales as λ^2 , even lightly doped semiconductor layers can contribute significant loss – hence THz QCL waveguides must be designed to minimize overlap with doped regions (except of course the doped active region layers which produce gain).

There have been several review papers in the past that have described the basics of THz QC-lasers and their challenges [28,59,60]. Here we will focus on advances in the past few years focusing on progress in: high-temperature operation, development of novel waveguides and cavities, and development of stabilized THz sources for spectroscopy.

3.1 High temperature operation: state of the art and new approaches

Despite over a decade of effort, unfortunately THz QCLs are still limited to cryogenic operation. The highest temperature operation for a QCL in the THz frequency range is currently 199.5 K in pulsed mode [61], but there have been a number of reports of lasers in the 160-190 K range [62,63]. These temperatures set an upper bound on continuous-wave operation due to the added issue of thermal management: ($T_{\text{max,CW}} = 129\text{ K}$ [64]). These lasers are based upon variations of the so-called resonant-phonon active region design [28]. This is somewhat similar to mid-IR active region designs, where rapid electron-LO-phonon scattering is used to depopulate the lower radiative state – although the details of the implementation differ considerably in the THz.

It is generally agreed that the major impediment to room-temperature operation for THz QCL is the reduction of gain at higher temperatures due to the onset of a thermally activated relaxation mechanism between the upper and lower radiative states based upon emission of optical phonons [28,65]. Since $\hbar\omega_{\text{THz}} < E_{\text{LO}}$, ($E_{\text{LO}}=36\text{ meV}$ in GaAs) at low electronic temperatures this scattering path is suppressed. However, at high temperatures electrons in the upper radiative subband gain sufficient in-plane kinetic energy to emit an LO-phonon and relax to the lower subband [28,65,66]. This process causes the upper state lifetime to decrease exponentially with increasing electronic temperature; this in turn reduces the gain and eventually leads to laser shutoff. It should be noted however, there are also minor effects which also contribute. The metal-metal waveguides become somewhat more lossy at higher temperatures as the Drude scattering rate increases [66]. Some simulations show broader gain linewidths (and reduced peak gain) at higher temperatures [67].

The most straightforward approach to improve the operating temperature is to explore novel active region designs within the GaAs/AlGaAs material system. Several designs have been demonstrated using an “indirect injection” scheme, where electrons are injected into the upper radiative state not by resonant tunneling, but by LO-phonon scattering from a higher energy state [68]. This scheme adds a degree of flexibility to design, and can in principle help to suppress sub-threshold parasitic leakage currents which plague THz QC-lasers. Good results have been obtained with this approach, particularly at lower frequencies – for example

150K operation at 2.7 THz [69], 163 K at 1.8 THz [70]. It seems likely that this scheme will be most useful for lasers < 2.5 THz where parasitic currents are the most problematic.

Given the lack of success in reaching room temperature operation using GaAs/AlGaAs based QCLs, other material systems become more attractive. There have been several THz QC-lasers demonstrated using InGaAs quantum wells with various barrier materials (InAlAs [71,72], GaAsSb [73], and AlInGaAs [74]). However none of these materials has matched the performance of GaAs/AlGaAs designs – likely due to the larger barrier heights, and perhaps increased alloy disorder scattering. Although InGaAs has a smaller effective mass than GaAs, which (in theory) increases the gain and reduces non-radiative scattering, these materials still have LO-phonon energies near 30–40 meV. Thus we might expect they will suffer from the same limitations of thermally activated phonon scattering as in GaAs-based wells. The use of III-nitride quantum wells is more promising; GaN has an LO phonon energy of 92 meV and hence thermally activated LO-phonon scattering should be dramatically suppressed at room temperature [75]. However, nitride quantum wells have proven to be very challenging to grow for vertical transport applications quantum cascade devices – even achieving reliable and repeatable RTDs is a challenge which has just begun to be solved [76,77]. Furthermore, the large built-in polarization fields for c-plane GaN creates sawtooth quantum well profiles. However, advances are occurring steadily: THz intersubband absorption has been measured in various step-well structures designed to flatten the band profile [78,79], and in quantum wells grown on the non-polar m-plane [80].

An even more radical approach is to suppress nonradiative scattering by implementing a cascade laser in quantum-dots instead of quantum wells [81,82]. The idea is based upon the so-called “phonon-bottleneck” mechanism: the zero-dimensional quantization in a quantum dot will eliminate electronic states at the LO-phonon resonance energy, which suppresses both relaxation and dephasing scattering. While this idea was originally proposed for mid-infrared QCLs (which have photon energies $\hbar\omega > E_{LO}$), it is in the THz where the payoff is greatest. Both density matrix [83], and non-equilibrium Green’s function theory simulations [84] predict significant gain present in THz QD QC-lasers at 300 K, although the transport physics are strongly altered due to the expected formation of strongly coupled electron-phonon polarons [85]. Efforts to use self-assembled quantum dots embedded into quantum wells have not been successful at demonstrating lasing but has led to renewed interest in the “top-down” etching of semiconductor nanowires [86,87]. This approach has the advantage of starting with high quality planar epitaxial material. However, it has its own challenges, including the difficulty of etching high aspect ratio nanowires with the required diameters of ~ 30 –50 nm. For example, a recent attempt found that lasing in a nanopillar THz QC-laser ceased when the etched pillar diameter fell below $5\ \mu\text{m}$ – likely due to surface damage/state induced depletion [88]. Because of these difficulties in achieving room temperature operation, there has been considerable effort to leverage the advances in high-power two-color mid-IR QC-lasers for intracavity THz difference frequency generation at room-temperature [89]. This approach has several appealing aspects, since the intersubband transitions within the QC-laser material itself provide the $\chi^{(2)}$ nonlinearity, the sources are monolithic and operate at room temperature, and wide fractional THz tunability can be obtained with only modest tuning of the mid-IR modes [35,36]. The highest reported output powers to date are 1.4 mW in pulsed mode and 3 μW in CW mode [36]. The challenges reside primarily in the conversion and overall wallplug efficiency, which is $\sim 10^{-5}$ or less. Coupling of the THz radiation out in a useable fashion is also a challenge, since free-carrier losses make the mid-IR laser cavity is quite lossy for THz. The most effective approach to date is a “Čerenkov” coupling scheme, where the THz radiation is coupled out in a distributed fashion in a cone-beam through the substrate [35].

3.2 Novel cavities for high power and beam shaping

A great deal of progress has recently been made on the difficulty of coupling of the generated THz radiation out of the QC-laser cavity in a high quality beam. The problem results from the

subwavelength nature of the preferred “metal-metal” waveguide. This involves the QCL gain medium (typically 5-10 μm thick) sandwiched in between metal cladding layers and etched into a laser ridge – similar in form to a microstrip or parallel plate waveguide. This waveguide has very little doped semiconductor cladding and allows scaling of the transverse dimensions to sub-wavelength dimensions – for example ridges as narrow as $\sim 15\ \mu\text{m}$. This is very advantageous for thermal dissipation – as a result metal-metal waveguides have been the only devices demonstrated with CW operation at temperatures $> 100\ \text{K}$ [64]. An interesting functionality is also made possible by the subwavelength nature of the metal-metal waveguide, when scaled to extremely narrow widths; for example, the laser cavity can be tuned over $\sim 300\ \text{GHz}$ by using a MEMS structure that interacts with the fringing field [89–91]. However, the subwavelength waveguide dimension has a disadvantage: the facet leads to a strong impedance mismatch of the waveguide mode with free-space, and acts as a point source with a highly divergent beam. Initial efforts to solve this employed a variety of solutions: facet-mounted lenses [92], horn antennas [93,94], 2nd-order DFBs and photonic crystals [95–97] – none of which simultaneously gave high output power combined with excellent beam patterns and CW operation over 77K. In the past five years however, several new and promising approaches have emerged.

First is the use of 3rd order DFB cavities, where the 3rd order diffraction provides optical feedback (for single-frequency operation). When the effective index of the waveguide is properly designed, the 3rd order diffraction couples into an end-fire antenna mode, where the beam divergence depends only on the length of the cavity, not on the transverse dimensions [98,99]. Hence narrow beams with divergence as low as 5 degrees from metal-metal waveguides have been reported. For example, this allowed Wienold *et al.* to operate a device up to 129 K in CW mode in a $15\ \mu\text{m}$ wide device – at 80K it maintained 0.6 mW level power. Research is active on how to increase the output power levels from 3rd order DFBs – one promising scheme is to increase the radiative aperture by integrating periodic slot antenna-like structures into the waveguide itself [100].

Second, the new approach of a “graded photonic heterostructure” is a modified form of a 2nd order DFB cavity designed to lase in the high radiative efficiency symmetric band edge mode. While this mode is present in every 2nd order DFB, usually a laser prefers to lase in the antisymmetric mode, since its anti-symmetric fields tend to cancel, thus dramatically reducing the radiative loss. However this leads to low output power. The photonic heterostructure designs engineer lossy boundary conditions so that the antisymmetric mode is selectively absorbed at the waveguide edges, so that the strongly radiating symmetric mode is preferred. This approach has demonstrated high powers from single mode lasers, such as over 100 mW power in pulsed mode at 10 K with high slope efficiencies of 230 mW/A [101], 25 mW of CW power at 10 K, and 1 mW of CW power at 80 K [102].

While these approaches are so far very successful for single mode lasers, they are all frequency-selective in nature, and so are not appropriate for broadband coupling needed for amplifiers, widely tunable lasers, and frequency combs. Hence, broadband, efficient integrated couplers still remains a challenge, particularly at very high powers. A possible option could be the post-process integration of an-index matched collimating lens. Another approach that has just begun to be explored is the use of quasi-periodic structures (1D gratings, 2D quasi-crystals) to break the symmetry of the radiating modes [103–105].

4. Applications

QCLs emitting in the mid and far IR spectral regions are very well suited for interrogation of molecules. Indeed, the mid-IR region (that can be roughly defined as the 2.9 - 24 micron wavelength interval) is the well-known molecular “fingerprint region,” where the strongest fundamental ro-vibrational bands, that allow to distinguish specific molecules and isotopomers, lie. In the far-infrared region, instead, rotational transitions can be excited for molecules possessing a permanent dipole moment. The intensity of such transitions can be

comparable with the strongest ro-vibrational transitions in the mid-IR. However, sources, components and detectors in the far-IR are far worse in terms of performance, cost and ease-of-use, also due to the need of cryogenical cooling (often at liquid-He temperature) for most lasers and best performing detectors. Present-day availability of THz-emitting QCLs (although not yet commercial) is partially filling this gap. In the following, we review recent applications of mid and far-IR QCLs to high-resolution spectroscopy and comb-assisted metrology.

4.1 Spectroscopy

The availability of compact, room-temperature operated QCLs, that are convenient single-frequency, relatively high-power, narrow linewidth laser sources, is matching increasing demand for spectroscopic applications encompassing environmental monitoring, security and biomedical sensing, smart sensor networks as well as more fundamental molecular studies and frequency metrology [30]. Several key advancements have favored applications of mid-IR QCLs, in the last two decades, as reviewed in paragraph 2.1. A key enabling feature for extensive spectroscopic application of mid-IR QCLs is the broad tunability, obtained mainly by use of an extended-cavity configuration [32,106] though, even before its demonstration, a number of spectroscopic techniques were developed. Among them we cite: standard direct absorption spectroscopy [107], modulation of wavelength [108,109] or frequency [110], differential or quartz-enhanced photoacoustic detection [111–113], polarization spectroscopy (that exploits birefringence induced in the gas by optical pumping) [114] or cavity ring-down spectroscopy, making use of high-finesse optical cavities [115].

The development of far-infrared (or THz) QCLs [27] more recently allowed extension of sensing technique across the far-infrared. This spectral range is very interesting for spectroscopy, due to the presence of rotational molecular bands that have absorption intensities comparable to those of ro-vibrational transitions. However, cryogenical cooling, often at liquid-He temperatures, is often required for sensitive detection, to cut-off the strong blackbody emission. Such a limitation, together with the still low-performing “optical” components available, halfway between microwave and optical technologies, has hindered a full exploitation of THz technologies. THz QCLs have partly filled the very scarce availability of coherent sources in this range, though cryo-cooling and very limited tunability have limited the number of spectroscopic applications. In particular, classical direct absorption spectroscopy [115], or commonly used schemes, like wavelength modulation [116,117] or differential spectroscopy [118], heterodyne spectroscopy [119] or quartz enhanced photoacoustic spectroscopy [120] have been implemented, although more complex schemes, which are commonly used in other spectral windows, have not yet been demonstrated.

4.1 Frequency metrology with QCLs

QCLs have gained acceptance for metrological frequency domain applications [121]. Their peculiar intrinsic low frequency noise was theoretically described at an early stage [122,123] but only recently was their intrinsic linewidth, (i.e. the quantum-limited frequency fluctuations or the Townes-Schawlow linewidth) has been unambiguously observed in the mid-IR [124,125] and in the THz range [126,127]. From these pioneering studies, it clearly emerged that an overwhelming contribution to frequency noise generally comes from the QCL current source. Therefore, low-noise current drivers have been developed to single out the QCLs elusive intrinsic noise. To such purpose, in the experiments reported to date, frequency domain fluctuations were converted to intensity fluctuations. This was more often done, by using the side of a Doppler-broadened molecular line as discriminator. From frequency noise spectral density curves obtained in this way, a quantum-limited linewidth of about 200 Hz for room-temperature operated mid-IR QCLs was found and less than 100 Hz for THz QCLs. Several experimental schemes have already been devised to exploit such

narrow linewidths for spectroscopic applications. They generally rely on phase/frequency locks to molecular lines [128], high finesse cavities [129] or other laser sources with a very low frequency jitter [130] or onto QCL emission control by injection of radiation from an external, stable source [131]. Although the main mechanisms determining such linewidths, that are the best for semiconductor lasers, have been unveiled, a number of unsolved questions still remain. Among these, a thorough understanding of the origin of QCL noise, in particular flicker noise, would pave the way to new laser designs for much less noisy QCLs, even better suited for demanding applications. Theoretical studies, going hand-in-hand with experiments, are trying to shed light on this key issue [132].

A very recent field of research on QCLs is the investigation of new schemes for referencing them to frequency comb synthesizers or also for generating comb-like radiation patterns directly from QCLs.

Referencing a QCL to a frequency comb synthesizer can provide frequency stability limited, in principle, by that of the comb itself and, therefore, by the clock used to stabilize the comb. While cesium fountains have a stability of about $10^{-14}/\tau^{1/2}$, the best optical clocks are even two to three orders of magnitude better. In addition, unlike molecular references, comb referencing allows to freely tune the wavelength of the QCL while reducing its frequency jitter and knowing its absolute emission frequency. Therefore, as early as in 2007, the first absolute frequency measurement, performed using a QCL at 4.43 μm referenced to an optical frequency comb synthesizer, was reported [133], using a frequency up-conversion scheme. More recently, a “smart” phase-lock to an optical comb was done with a mid-IR QCL that provided a full tunability and a linewidth below 1 kilohertz in a 1 ms timescale [134]. This also allowed scanning the laser around a carbon dioxide molecular transition obtaining an overall uncertainty of 1 kHz in the frequency determination (about 10^{-11} fractional uncertainty). Also for THz QCLs, referencing to a near-IR comb [135] was shown. More recently, direct referencing of a QCL, emitting around 2.5 THz, to an air-propagating THz comb was obtained [136], thus enabling spectroscopic scans over molecular lines with an absolute frequency scale providing an uncertainty of a few parts in 10^{-11} on the laser frequency and an uncertainty on the line-center determination of 10 kHz [137]. The recent demonstration of direct comb emission from mid-IR and THz QCLs represents a compact alternative to referencing to external combs [44,47,48] although absolute frequency referencing is not provided. Here it is not a mode-locked train of pulses in the time domain that gives rise to the comb spectrum in the frequency domain. Also no conversion up-conversion processes are involved [138]. Instead, a multimode emission with a controlled dispersion within the active medium, combined with four-wave mixing, generates the comb-like spectrum. Although metrological-grade applications still need to be demonstrated, these first results and the recent advent of octave spanning THz QCLs [139] are quite encouraging for direct comb generation in QCLs.

5. Focus issue

In this focus issue of Optics Express are reported some of the latest advances and developments in the field of quantum cascade lasers, in the 20 year anniversary of this groundbreaking device, presented during the International Quantum Cascade Laser School and Workshop (IQCLSW 2014), held in Policoro (MT), Italy.

In the spotlight there is the understanding of the physical mechanisms governing the operating regime of Mid-IR and THz QCL optical frequency combs (OFC). The careful analysis of four-wave mixing in broadband gain, low dispersion mid-IR cavities underline future routes for broadband QCL frequency combs (Villares *et al.*); furthermore through shifted wave interference Fourier Transform Spectroscopy (Burghoff *et al.*) elucidate the time domain properties of THz QCL-OFCs, demonstrating frequency and amplitude modulation signatures.

The remarkable performance of Mid-IR QCLs nowadays capable to deliver continuous wave power output up to 5.1 W at room temperature, in a broad spectral range from 3 to 12 μm , with monolithic sampled grating design and on-chip beam QCL combiner are critically discussed in the manuscript of Razezghi *et al.*, which also elucidates the advantage of such a major improvement, for the demonstration of room temperature THz QCLs providing up to 1.9 mW peak power on a widely tunable 1-5 THz frequency range. State of the art performance levels on both the long-wavelength side of the mid-IR window and the low frequency boarder are discussed by Bahriz *et al.* reporting up to 80°C QC lasing at 20 μm . The research on integrated solutions for spectroscopic applications has led to innovative devices like spiral cavity superluminescent (SL) emitters at 5 μm providing 57 mW of SL power at 250 K with a 107 μm coherence length (Zheng *et al.*) and to broadband (5.9 μm - 7.2 μm) superluminescent emitters (Riedi *et al.*) of potential use as amplifiers for mid-IR frequency combs. Progress in the field of low consumption devices for integrated systems are presented in the manuscript of Bismuto *et al.*, showing QCL optical powers larger than 20 mW with less than 1W electrical consumption. The versatility of mid-IR QCLs for down-conversion processes in the telecom range is discussed in the manuscript of Houver *et al.* demonstrating optical sideband generation in MIR QCLs up to room temperature, providing a simple method of stabilizing a QCL to an optical-comb through MIR up-conversion.

As a novel possibility the intersubband transitions, core idea of the QCL, can be successfully used to produce detectors matching the spectral range of the QCL. Reininger *et al.* report on a resonant cavity quantum cascade photodetector promising a 95mA/W responsivity and a specific detectivity of $\sim 10^9$ Jones at T = 300K.

Such a major improvement opens the path to possible on-chip detector/source integration for an all-intersubband high performance sensing platform, as discussed and demonstrated by Jagerska *et al.* With an alternative approach Krall *et al.* demonstrate simultaneous on-chip generation and detection of THz radiation in coupled cavity systems, using a single semiconductor heterostructure, based on a four-well resonant phonon THz QCL. It is clear that the beam divergence here plays a very fundamental role. Monolithic solutions targeting the improvement of the optical beam profile of THz QCLs are also discussed in the issue presented by Halioua *et al.* reporting on phase-locked arrays of surface emitting THz QC lasers with 10° degree optical beam divergence and by Castellano *et al.* discussing on a novel micromachined feed-horn waveguide, assembled around a double-metal THz QCL, and integrated with a slit-coupler providing a 20° beam divergence. The possibility to employ THz QCLs as powerful and broadband sources for time-domain systems promising to boost the signal-to-noise ratio is now a concrete perspective. Bachmann *et al.* demonstrate a heterogeneous broadband QC based THz amplifier that employs an integrated source of coherent THz radiation, discussing the potential of this novel technology. Pushing the performance of THz QCLs at a higher level of maturity in terms of CW, RT and wall-plug efficiency potential is related to a careful compromise between bandgap engineering and high-quality molecular beam material growth as discussed by Li *et al.* Despite the present limitations, the QCL technology has already a prominent impact on sensing applications as extensively discussed in the present focus issue. Zhao *et al.* propose a broadband THz sensor based on arrays of single-mode QCLs, promising the possibility to probe the complex refractive index of an analyte on a broad range of THz frequencies; the potential of V-shaped and ring-shaped THz resonant cavities is presented by Campa *et al.* envisioning their impact for the next generation of high-sensitivity and high-resolution QCL-based THz spectroscopic systems, promising build-up cavities and novel frequency references for QCLs; high optoacoustic transduction efficiency set-up for quartz enhanced photoacoustic spectroscopy (QEPAS) in the THz are presented by Spagnolo *et al.*, underlining this potential of the QEPAS technique for highly selective gas recognition in the infrared; the potential of external cavity mid-IR QCLs for stand-off detection of broadband absorbers is discussed by Macleod *et al.*, employing a widely tunable active coherent laser spectrometer; alternative technologies

based on mid-infrared QCL-coupled silicon-on-sapphire ring resonator gas sensor are presented and discussed by Smith *et al.*, offering a concrete perspective of size-shrinking for trace-gas detection systems; the potential of combined QCL-tunable laser diode systems for simultaneous measurements of distinctive isotope ratios is discussed by McManus *et al.*

The potential of the QCL mostly comes from its “*quantum engineering*” design strategy. Driving these devices in new operating regime means also to find solid design and engineering strategies enabling to predict the device behavior and to improve its functionalities. In this focus issue, some recent modeling progresses in the several different contexts are described: i) predictions of the “dissipative quantum transport” by developing new approaches and simulation strategies (Greck *et al.*), ii) engineering of the waveguide structures via a rigorous theoretical test, that takes into account increased waveguide loss, Auger recombination, and Purcell enhancement of spontaneous recombination (Khurgin); iii) Monte Carlo analysis of the terahertz difference frequency generation susceptibility to consolidate the development of RT THz sources (Jirauschek *et al.*); iv) limits and prospects for pushing QCL operation in the mode locking regime (Jung *et al.*); v) detailed analysis of several competing models for interface roughness scattering (Lindskog *et al.*).

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