Invited Paper

QCL based terahertz frequency metrology

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Abstract: Quantum cascade lasers combine unique features in the framework of THz sources, namely compactness and *mW* output power. In addition, owing to an intrinsically low phase-noise level, they are suitable for frequency and phase stabilization, which makes them attractive sources for frequency metrology applications, and able to overcome the power limits of current microwave-traceable THz emitters. In this contribution, we address metrological grade THz quantum cascade lasers from their intrinsic spectral feature to the use of optical frequency combs as frequency reference.

Keywords: Terahertz generation, Coherent detection, Frequency metrology.

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1. Introduction

Quantum cascade lasers (QCLs) are unipolar devices that exploit optical transitions between electronic states (conduction subbands) arising from spatial confinement in semiconductor multi quantum wells. By means of hetero-structure engineering, the energy of such transitions can be tailored so as to cover a very broad spectrum, resulting in laser emission from the near to the far infrared (IR) [1]. In the mid-IR region, where they were formerly demonstrated, QCLs are at present mature commercial devices, which operate in CW at room temperature [2-4], with watt-level output power [3, 5, 6], and can be tuned by up to a few microns when being included in an extended-cavity [7, 8]. On the other hand, design and fabrication of QCLs at terahertz frequencies (i.e. for energies below the optical phonon energy) has revealed more difficult: due to the low photon energy. Achievement of the population inversion is challenging, especially at room temperature ($h\nu < KT$), while waveguiding is made difficult by the large wavelength value ($\approx 100\ \mu m$) and is affected by free-carrier absorption. As a consequence, after ten years since the former demonstration of a THz QCL [9], these lasers can still operate only at cryogenic temperature, up to 200 K in pulsed mode and 120 K in continuous wave (CW), with output power in 100 μWs-10 $s\ mW$ range [10-12].

The terahertz spectrum, i.e. the frequency region ranging from 0.1 to 10 *THz*, is well known to be an underexploited window of the electromagnetic spectrum. Due to the lack of efficient compact sources and sensitive detectors, practical applications of THz radiation, which are numerous and attractive, are still mainly confined to laboratories. Despite their present need of cryogenic cooling, QCLs are thus considered among the most promising sources of THz radiation, mainly because of their compactness, relatively high output power, and covering of the whole spectral region between 1.2 and 4.9 *THz* [13, 14]. QCLs have proved to be effective sources for eg THz imaging, which is one of the most appealing applications of THz radiation, since many materials that are transparent at THz frequencies are opaque in the visible spectrum, and vice versa. Traditionally, THz imaging is performed using low power (*nW*–*μW*) pulses generated by rectification of femtosecond optical pulses incident on a photoconductor or nonlinear crystal [15]. Images are then acquired by rapidly scanning the sample or by means of a fast steering mirror [16]. On the other hand, THz imaging systems based on QCLs and focal plane array microbolometric cameras have demonstrated real time acquisition capability, thanks to the high available optical power [14, 17].

QCLs can also find applications in molecular spectroscopy, the traditional applicative domain of terahertz radiation. THz molecular spectroscopy has an amazing scientific potential, since many absorption and emission molecular lines of interest in astrophysics and atmospheric sciences fall in this spectral region, where many chemical species have very strong characteristic rotational and ro-vibrational transitions [18]. In particular, for gases, the typical absorption strengths are 10^3–10^6 stronger than in the microwave region [19]. In order to investigate the structure and the energy levels of molecules and measure line broadening and frequency shifts, it is crucial to dispose of tunable sources with high resolution, as high as $\Delta\upsilon/\upsilon \approx 10^{-6}$, and high accuracy. To address such requirements, several technologies providing tunable THz emission have been developed in the last decades. In this respect, frequency conversion techniques from both the electronic and the optical frequencies have proved more convenient so far than direct THz emitters, such as backward wave oscillators or molecular gas lasers: i) up conversion, or the harmonic generation from microwaves sources, has been successfully exploited for many years: a high power (100 s *mW*) microwave source in the 10-100 *GHz* range is typically up converted by means of cascaded frequency multipliers, such as Schottky barrier diode mixers, resulting in *μW* output power up to 2-3 *THz* [20]; ii) well established photomixing of two near-IR tunable laser oscillators is usually performed by means of a low-temperature grown GaAs or InGaAs photomixer, and allows coverage of the THz spectrum of up to 2-3 *THz*, with an emitted power in the 10 *s nW* to the 10*s μW* range [21]. It is worth to notice that the down-converted radiation can be straightforwardly referenced to a microwave frequency standard by locking the two near-IR lasers to an optical frequency comb [22].

The above tunable sources are in general complex and/or expensive, while the emitted THz power stays well below 1 *mW* above 1 *THz*. Recourse to more powerful QCLs would thus result in a significant gain in terms of sensitivity, and would give access to power demanding experiments, such as saturation spectroscopy measurements. Concerning frequency resolution, spectral features of THz QCLs were first studied through the use of mixing techniques [23, 24], which have pointed out a linewidth of the order of ~10 *kHz* on a ms timescale, limited by temperature and driving current fluctuations. On the other hand, due to a small expected linewidth enhancement factor (LEF) arising from their symmetric gain spectrum, THz QCLs should exhibit intrinsic linewidths in the sub-kHz range [25]. These features were firstly demonstrated by measuring the LEF of a 2.6 *THz* QCL, using a self-mixing technique, which leads to an estimated LEF of ~0.5 [26]. Such a low frequency noise is favorable to active linewidth reduction through both frequency- and phase-locking techniques [27]. Indeed, several demonstrations of frequency stabilization have been reported so far [28-34], with the aim of obtaining a high power, metrological grade THz source, possibly referenced to a microwave standard, which would overcome the power limits of current microwave-traceable THz emitters.

In this contribution we address metrological grade THz quantum cascade lasers and their possible applications. The paper is organized as follows. In the first part we discuss the spectral features of THz QCLs and review a few phase noise measurements carried on in the last years. Then we will address coherent detection of THz QCLs, based on the different kind of local oscillators and detectors. In this respect, we will give an overview of a novel generation of fast FET detectors that hold a high potential as THz frequency mixers. In the last section, we review the state-of-the-art of frequency- and phase- stabilized THz QCLs, and discuss their use for high resolution spectroscopy.

2. THz QCL intrinsic linewidth

The intrinsic linewidth of a laser is ultimately related to the uncertainty principle of quantum mechanics, and can give very important information on the physical limits of the spectral purity of the laser source. Measuring the intrinsic linewidth of a THz QCL would therefore mean determining its achievable spectral resolution and coherence length, and thus understanding whether QCLs are suitable for metrological grade applications.

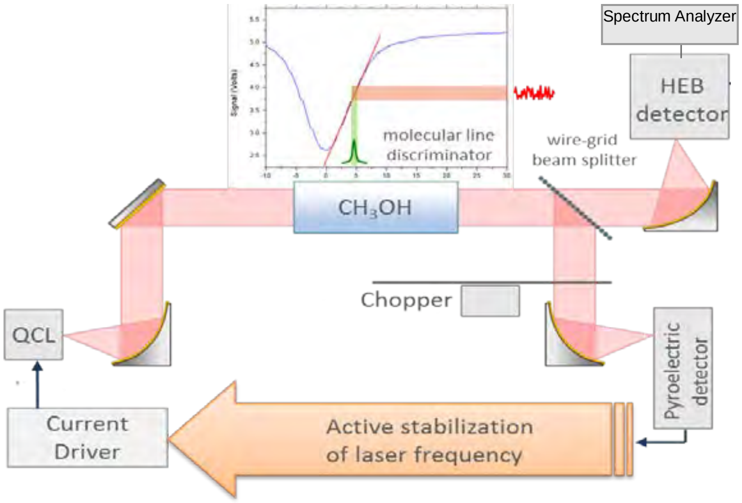

Fig. 1 Schematic of the experimental set-up for the THz QCL intrinsic linewidth measurement. The discriminator function of the absorption molecular line is shown, in order to convert frequency fluctuations into detectable intensity fluctuations.

One possible way to measure the spectral purity of a THz QCL is to measure its frequency-noise power spectral density (FNPSD), providing an experimental evaluation of its intrinsic LW. In [35] it has been shown how detectable intensity (amplitude) variations can be used to retrieve information on the laser frequency fluctuations, by simply means of a discriminator. In [36] a very similar procedure has been followed for a THz QCL, using the side of a Doppler-broadened methanol molecular transition as a discriminator.

As shown in Fig. 1, the THz radiation coming from a QCL is collimated and sent through a spectroscopy cell filled with methanol vapours. The optical beam is then split into two parts: the first one is sent to a Pyroelectric detector, while the second part is delivered to a faster detector such as Silicon bolometer or Hot Electron Bolometer (HEB). Scanning the laser current, its frequency is swept across a methanol absorpiton, specifically the ro-vibrational molecular transition line of CH_3OH, centered at $\upsilon_o = 2.5227816\ THz$, and it is detected by means of the pyroelectric detector. In this way not only one side of the transition can be used as discriminator, but the laser frequency can be stabilized at the half height of the slope by means of a slow (<10 *Hz*) software PI loop circuit acting on the laser current. Moreover in this way the QCL behavior at higher frequencies is not affected. At the same time, the second part of the beam is acquired by means of the bolometer and used for the frequency-noise measurement. Given the intrinsic low-noise nature of the measurement, the discriminator must introduce a negligible noise providing, at the same time, a gain factor suitable for a good detection.

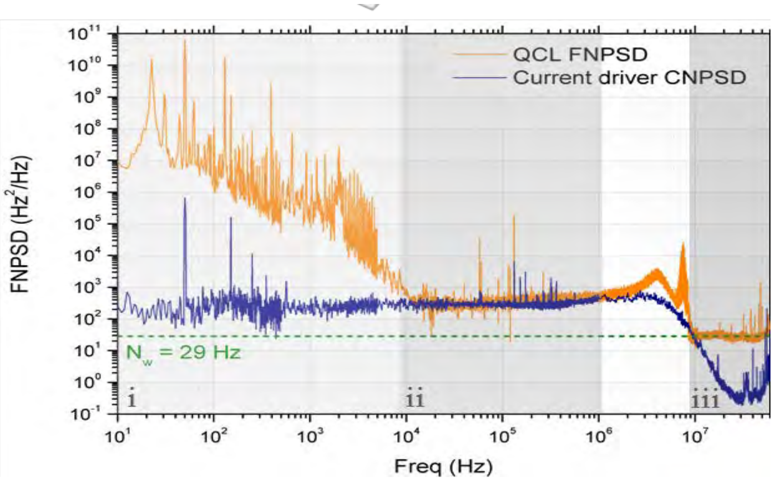

Fig. 2 Experimental FNPSD of the THz QCL (orange trace), compared with the contribution to frequency-noise of the CNPSD of the current driver (blue trace). The dashed line marks the white noise level.

The final result is shown in Fig. 2. This technique allows a measurement of the FNPSD of the QCL over seven frequency decades (from 10 *Hz* to 100 *MHz*) and 10 amplitude decades. In order to better understand it we have plotted on the same graph also the current-noise power spectral density (CNPSD) of the current driver, converted to the same units by using the current tuning coefficient. We can observe three different frequency domain: i) At frequencies up to 10 *kHz,* the FNPSD is dominated by a noise not due to the laser driver, but arising from the QCL itself. ii) In the range from 10 *kHz* to 5 *MHz,* the CNPSD is the main cause of fluctuation in the laser frequency, therefore the FNPSD is dominated by current laser fluctuations. iii) At frequencies above 8 *MHz*, the FNPSD flattens to a value significantly different from the CNPSD, leading us to the measurement of the white noise level N_w. From this value we can easily extract the QCL intrinsic linewidth as $\delta\upsilon = \pi N_w = 90 \pm 30$ *Hz* (FWHM).

This has been the first measurement of the intrisic linewidth of a THz QCL intrinsic LW, but it has been supported by theoretical calculations, that give a value of $\delta\upsilon = 107$ *Hz*, and by more recent experiments [37], with a value of $\delta\upsilon = 230$ *Hz*.

3. Coherent detection

Despite their high output power, detecting the radiation emitted by THz QCLs remains an unsolved problem. To date, most sensitive detectors are composite Si-bolometers or super-conducting HEBs [38, 39], which both require cooling at Liquid-He temperature. Room temperature detectors, such as pyroelectric detectors, schottky diodes, and golay-cells are either slow or scarcely sensitive. This represents a clear drawback, which has so far hindered the exploitation of THz QCLs. An alternative approach is represented by heterodyne detection schemes, which can give access to fast response and high signal dynamics at room temperature.

In addition, such techniques can be exploited for frequency- or phase-stabilization of the THz QCL to the local oscillator, so as to narrow the QCL emission line, which is generally much broader than its quantum limit, due to temperature and driving current fluctuations.

Heterodyne detection of THz QCLs was firstly performed by using diode mixers and a FIR gas laser or a second THz QCL as local oscillator [24, 40]. A few years later, both Khosropanah et al. and Rabanus et al. demonstrated the use of microwave local oscillators up-converted in the THz range, in combination with a hot-electron bolometer (HEB) mixer [30, 31], a solution which also allows directly referencing the QCL frequency to a microwave frequency standard. All of these detection techniques suffer of poor tunability and exhibit a limited signal-to-noise ratio, due to either the high noise-equivalent-power (NEP) of the diode mixer, or the very low power of the local oscillator. Both these issues were addressed by Barbieri and coworkers, who have introduced an original and effective mixing scheme based on the use of a femtosecond near-IR laser as local oscillator [32]. Such approach is inherently broadband and features a NEP of few pW in 1 *Hz*, comparable to that of liquid He cooled bolometers. Recently, Consolino et al. exploited a free-space THz comb local oscillator and HEB mixer, which yield a NEP as low as 100 *fW/Hz* [34].

In this context, fast field effect transistors (FET) operating at room temperature hold potential as both highly sensitive direct detectors and frequency mixers. Indeed, while most FET detector architectures, such as high-electron-mobility transistors and complementary metal-oxide semiconductor [41, 42], can operate only in the hundred of GHz range, the recent introduction of semiconductor nanowires (NWs) as the FET active channel has allowed efficient detection well above 1 *THz* (Fig.32). Owing to reduced dimensionality, high electron mobility at room temperature (≈1000 $s\ cm^2 V^1 s^{-1}$ for InAs NWs) [43] and efficient coupling of the incoming radiation by means of a THz antenna, a responsivity ≈12 *V/W* and a NEP as low as 100 $pW/Hz^{1/2}$ were first demonstrated at 1.5 *THz* [44]. This result has paved the way for the use of NW FET as detectors for THz QCLs, and such impressive performances have been since then extended up to 2.8 *THz* [45].

Besides proving attractive power detectors for THz QCLs, NW FETs are expected to have a response fast enough for their exploitation as frequency mixers, due to their small dimensions and thus low parasitic capacitance. To date, the 3 *dB* bandwidth of NW FETs was found to be as high as 100 *kHz*, limited by the parasitic capacitance of the detector package (~ pF), which is orders of magnitude higher than the intrinsic capacitance of the transistor (~ aF) [46]. Whereas 100 *kHz* is already much larger than the bandwidth of room temperature THz detectors, such as Golay cells and pyroelectrics, this leaves room for further future improvements and performance of THz QCL frequency mixing.

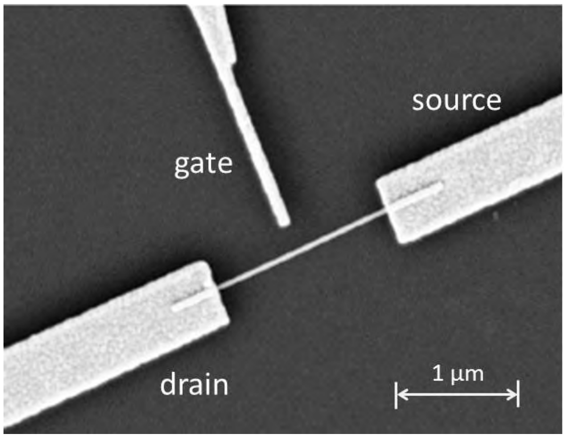

Fig. 3 Scanning electron microscope picture of an InAs NW FET.

4. Metrological-grade THz QCLs

THz QCLs have shown an exceptionally small intrinsic linewidth, which means that they are suitable for metrological grade applications. In fact their frequency/phase can in principle be stabilized against some secondary frequency standard. The first attempts in these direction were performed using molecular transitions, as for example in [28] where phase-lock of a 3 *THz* QCL at the 3.1059368 *THz* line of a methanol gas laser was demonstrated or in [46], where frequency stabilization of a single-mode THz QCL against the 2.409293 *THz* line of a CH_2DOH gas laser was obtained. In both cases, a narrowing of the beat signal was observed (65 kHz and 3 *kHz* respectively).

Another viable solution is to lock a QCL to a microwave-driven harmonically generated THz sources. This was successfully demonstrated at 1.5 *THz* [31], at 2.7 *THz* [30] (phase-locks), and at 2.3 *THz* [50] (frequency-lock). Despite the high complexity, the high electrical power consumption and the very low efficiency (with a few pW radiation power), such a kind of sources provide a very narrow and absolute-frequency reference.

Phase-locking a QCL to an OFCS was achieved only recently by Barbieri and co-workers by locking a 2.5 *THz* QCL to the nth harmonic of the repetition rate of a mode-locked erbium-doped fiber laser by using electro-optic detection [32] or a photoconductive antenna [33]. These two approaches take advantage of a room-temperature detection of the beat-note, but are both based on low-efficiency up-conversion processes, and thus require a CW THz power in the *mW* range.

This limitation can be overcome by moving to a native THz detection, where the beat-note can be acquired by a square-law THz detector. This will highly increase the detection efficiency, therefore involving only a small fraction of the overall emitted QCL power. In this case, a free-standing air-propagating THz comb is needed. Although all the commercial pulsed THz sources used in time domain spectroscopy have an intrinsic comb nature [51, 52], no direct use of such sources as frequency "ruler" for a THz QCL has ever been reported, until very recently. In fact in [34], phase-locking of a single-frequency CW QCL emitting at 2.5 *THz* to a single tooth of an air-propagating THz comb was demonstrated: direct knowledge of the QCL frequency and narrowing of its emission could be observed.

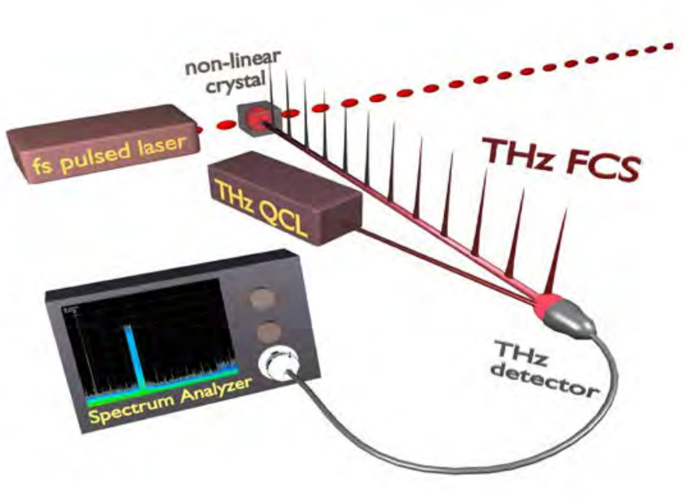

Fig. 4 Operational principle of referencing a THz QCL to an air-propagating THz comb.

The principle of the THz comb generation and the beating with the THz QCL are shown in figure 4. A femtosecond mode-locked Ti:Sa laser is focused in a single-mode waveguide fabricated on a MgO-doped $LiNbO_3$ crystal plate, in order to generate through optical rectification in Cherenkov configuration, the train of THz pulses of the comb. The obtained FCS is, in the frequency domain, a series of very narrow, equally spaced emitters, which cover the whole spectrum from 100 *GHz* to 6 *THz*. Moreover since the pulses are identical, the comb-like spectrum of the infinite train has a perfectly-zero offset, and the spacing corresponds to the 77.47 *MHz* repetition rate of the pump laser.

This repetition rate has been stabilized against a Rb-GPS (Global Positioning System) disciplined 10-*MHz* quartz oscillator, down to the mHz level, thus ensuring a stability of each tooth of the THz comb at the 100 *Hz* level. Using a HEB detector, with a very low NEP and high detection efficiency, it is possible to directly observe the beating signal between a very small fraction (about 100 *nW*) of the CW THz QCL and one tooth of the THz FCS.

The beat note detection gives the possibility to continuously trace the QCL frequency, but in order to effectively stabilize the phase of the QCL emission to the frequency comb reference, a phase-lock loop has been implemented. The electronics used in this case is quite standard: the beat-note signal is mixed with a synthesized fixed frequency and processed by an analog/digital phase detector. The correction signal closes the PLL on the fast (1 *MHz* bandwidth) modulation input of the QCL current driver. Figure 5 shows the beat note detected with the open (grey) and closed (blue) phase-lock loop.

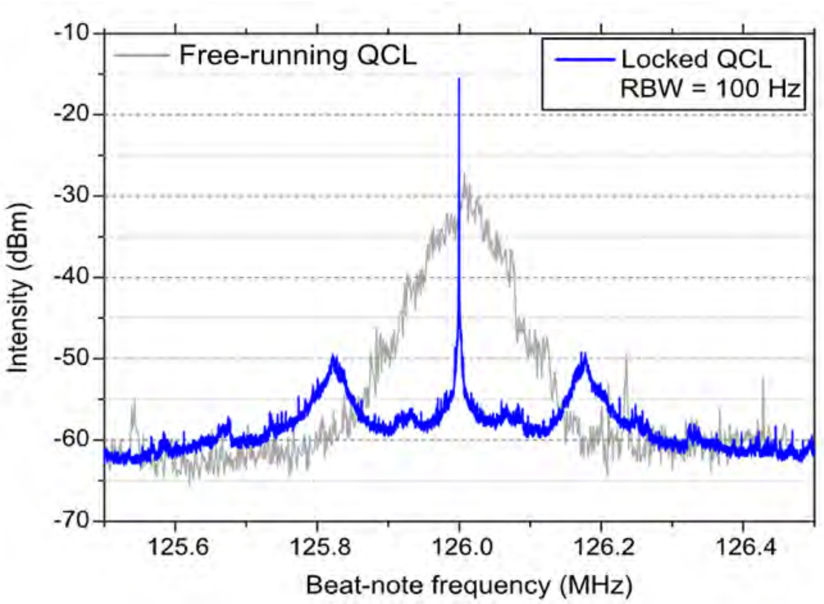

Fig. 5 Detected beat-note spectrum with a free-running (gray) QCL, and beat-note figure when the phase lock loop is closed (blue).

5. Conclusions

Combined use of QCLs and optical frequency combs is a promising approach for a metrological-grade investigation of THz spectrum. THz QCLs ensure coverage of the whole 1-5 *THz* frequency range with *mW* power level, and their intrinsic linewidth is favorable to line-narrowing by phase-locking to a stable oscillator. The use of a frequency comb reference provides a low-noise coherent link to the microwave range, resulting in extreme narrowing of the QCL emission and possible referencing to a frequency standard. Among the frequency mixing techniques developed to this aim, the highest sensitivity is presently obtained through the use Liquid-He cooled HEB mixers. However, recent development of low-noise fast electronic THz detectors based on semiconductor NWs will soon ensure a significant improvement of the sensitivity of room temperature mixing schemes.

Among the several possible applications of metrological grade THz QCLs it is worth mentioning the development of comb-assisted THz sub-Doppler spectrometers and of absolutely-referenced local oscillators for heterodyne THz spectrometers. Regarding the latter topic, in particular, not only the narrow linewidth of the local oscillator but also, and mostly, the stability over long time periods of its absolute frequency can represent a real breakthrough. A number of demanding new applications will be possible to afford, e.g. spectroscopic interrogation of cold molecules (see eg. [53]) as well as precise measurement of the long-term variation of fundamental physical constants from astronomical observation [54, 55].

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