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Quantum cascade laser: a compact, low cost, solid-state source for plasma diagnostics

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ABSTRACT: Quantum cascade lasers (QCL) are unipolar injection lasers based on intersubband transitions in a modular semiconductor heterostructure. The first THz QCL, operating at 67 μm (4.3 THz), was demonstrated in 2002; the wavelength range now extends beyond 250 μm (1.2 THz) and is entering the sub-terahertz frequency range for devices operated in external magnetic field. Although a number of different quantum designs have been demonstrated, increasing the operating temperature remains a major challenge: the maximum temperature is still $\sim$195 K, and recently approached 225 K in high magnetic fields. Nevertheless, compact continuous wave systems operating within Sterling coolers already ensure ample portability and turn-key operation and QCLs represent then the THz solid-state radiation source that actually shows the best performance in terms of optical output power, which can reach more than 100 mW average, and linewidth, typically in the tens of kHz for single mode devices. THz QCLs have then a realistic chance to deeply impact technological applications such as process monitoring, security controls, and bio-medical diagnostics. They are ideally suited though for plasma polarimetry and interferometry, thanks to their high polarization selectivity, excellent stability and ruggedness, and ease of high-speed modulation. Their compact size and monolithic cavity arrangement allows placement in the very proximity of the plasma to be monitored, easing requirements of stability against vibrations etc. Furthermore, the long coherence lengths should be easily compatible with interferometric arms of even very different lengths, a geometry ideal for coupling to a plasma reactor. The possibility of direct current modulation at MHz if not GHz frequencies ensures then an excellent temporal resolution of the meaurements, and a large low-frequency noise rejection. New analysis schemes also become feasible, for instance employing two-color lasers, operating at the same time at two appropriately

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chosen wavelengths, or exploiting the laser emission tunability for implementing frequency modulation techniques. This paper will discuss the current state-of-the-art in THz QCL technology and applications, focusing on those aspects of greatest relevance for plasma diagnostics.

KEYWORDS: Plasma diagnostics - interferometry, spectroscopy and imaging; Detection of explosives

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1 Introduction

A semiconductor heterostructure consists of a layered sequence of two or more semiconductor materials with different band-gaps. The resulting band-edge discontinuity provides an effective potential profile (with wells and barriers) for the electronic motion. If the layer thicknesses are of the order of the electron wavelength, quantum confinement splits the bulk bands into a number of individual subbands. Inter-subband optical transitions then become possible, for instance involving only electrons belonging to the first conduction band. The transition energy is mainly determined by the width of the quantum well where the electron wavefunctions are contained.

The idea of constructing a unipolar semiconductor laser operating on inter-subband transitions is due to Kazarinov and Suris and dates back to the seventies [1]. The concept was immediately appealing because of the artificial nature of the transition, which allows a full control by design on its properties (energy, dipole matrix element, non-radiative scattering rate, etc.). Furthermore, two fundamental aspects are particularly relevant for the implementation of a laser. Both subbands involved originate from the same bulk conduction band and therefore are ideally characterized by the same effective mass. This produces a nearly delta-like joint density of states, maximizing the peak gain and reducing its dependence on temperature. Moreover, at the end of the photon emission process, the electron is still in the conduction band, and can then be recycled in a subsequent identical active region to emit another photon, for a number of times equal to the number of periods.

Despite many practical proposals and attempts, the first inter-subband laser, emitting in the mid-infrared at 4.3 μm, was realized only in 1994 [2], at Bell Laboratories, thanks also to the development of modern semiconductor growing techniques, like molecular beam epitaxy (MBE). Population inversion was achieved through current injection via resonant tunnelling, and the device was named *quantum cascade laser*, reflecting the periodic potential profile with the above-mentioned "cascade" of transitions in repeated identical active regions.

In ten years, research on quantum cascade lasers (QCL) has rapidly developed, and the devices have reached considerable performance levels. They cover the whole mid-infrared range (from below 3 μm up to 25 μm); single-mode and tuneable emission is routinely achieved using distributed feedback (DFB) and external cavity resonators; continuous wave output powers at the Watt level have been demonstrated even at room temperature. Lately far-infrared QCLs with emission frequencies in the THz range (down to ~ 1 THz, and below in magnetic field) have also been realized, thereby effectively bridging the gap between quantum photonic sources and classical electronic oscillators [3].

The availability of a high-power solid-state far-infrared laser source could deeply impact the progress of laser-based techniques for plasma diagnostics. Polarimetry and interferometry measurements in this frequency range, in fact, usually rely on bulky, optically-pumped gas lasers, requiring rather complex set-ups, stabilization, etc. Reaching output powers of the order of 100 mW and above, and displaying highly polarized emission with long coherence lengths of several meters, THz QCLs seem then a possibly ideal solution to replace present source technologies. With the intrinsic compactness and power stability, not to mention low cost, of a semiconductor laser, they could indeed be placed in close proximity of the plasma reactor, reducing vibration and encumbrance issues, at the same time offering the capability of easy electrical modulation, which can be performed at much higher rates than through mechanical choppers. This latter aspect should be beneficial for increasing temporal resolution and eliminating problems related to low-frequency noise. Furthermore, by appropriate design of the structure, QCLs can offer functionalities normally precluded to conventional gas lasers. For instance they can be engineered to emit at two well distinct wavelengths at the same time, or they can be operated in a self-mixing configuration in which the output beam is re-injected into the laser, made to interfere with the internal field, and detected directly in the device electrical response.

2 THz quantum cascade lasers: design and waveguiding

The extension of the QC scheme to photon energies below the reststrahlen-band was not straightforward. The task for the quantum designer is in principle the same as for the mid-infrared: achieving a long lifetime of the upper laser level and a fast depopulation of the lower laser level, while maintaining a decent optical dipole matrix element between the two states. But in this frequency range, the use of resonant longitudinal-optical (LO) phonon scattering processes to engineer the lifetimes of each level, and thus the population inversion, is rather limited, because the two radiative levels are separated by a small energy, and it is thus difficult to efficiently depopulate one without also affecting the other. The first THz QC designs mainly focused on engineering a long lifetime of the upper level, by keeping the energy difference between the levels small enough to avoid LO-phonon emission. Using superlattices for the active region, it was shown that population inversion can be achieved by employing electron-electron scattering within the lower miniband of the superlattice. The dispersion of this miniband is designed to be as large as possible to avoid thermal backfilling, but smaller than the photon energy in order to avoid reabsorbtion [see figure 1(a)]. This, together with a new waveguide concept, led to the first demonstration of a terahertz quantum cascade laser emitting 2.5 mW at 4.4 THz in pulsed mode, with a maximum operating temperature of 50 K [3]. Following this initial work, performance increased rapidly, with the demonstration of continuous wave (cw) operation [4], maximum lasing temperatures up to 195 K [5], output powers up to 250 mW [6], and an emission frequency down to 1.2 THz [7]. Alternative design concepts for the active region were indeed necessary to improve performance. Higher temperatures were achieved

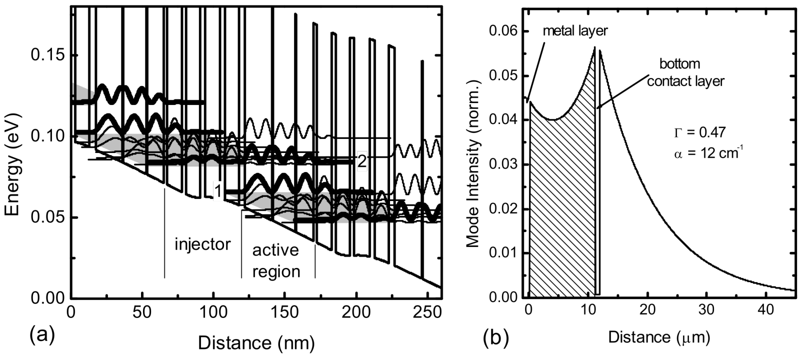

Figure 1. (a) Conduction band energy diagram and moduli squared of relevant wavefunctions of the first 4.4 THz QC laser under an electric field of 3.5 kV/cm. The optical transition occurs between the two states drawn in boldface. Carriers are injected from the ground state into the upper laser level via resonant tunneling. (b) Calculated waveguide mode profile along the growth direction of the final device structure. The origin of the abscissa is at the top metal-semiconductor interface; the laser active core is indicated by the shaded area.

with the re-introduction of the phonon depletion scheme: direct electron-phonon scattering was applied to depopulate the lower laser level, but, unlike in mid-infrared QCLs, the two levels in resonance with the LO-phonon were not placed in the same QW as the radiative transition [8]. The two separate sections were instead appropriately tunnel-coupled, so that extraction through phonon emission could apply selectively mainly to the lowest subband of the lasing transition. An important concept for higher output powers and lower emission frequencies was also the bound-to-continuum design [9], which addresses the problems of efficient injection into the upper level at low transition energies while maintaining favorable transport properties.

From the waveguide perspective, already QC lasers in the long mid-infrared range used a combination between a surface plasmon (SP) and dielectric confinement: a low refractive index material is grown below the gain medium, whereas the metallic layer atop creates a SP, the two different claddings leading then to a tightly confined TM mode peaked at the metal interface. As the wavelength is further increased into the THz, however, the layer of low refractive index is no longer sufficient to confine the mode. In the first THz laser, the latter cladding was therefore replaced by a thin highly-doped semiconductor, and the entire structure was grown on a transparent semi-insulating (SI) substrate [3]. In this way, another surface plasmon layer at the bottom of the active region is created, which also provides the electrical contact. Its doping and thickness are optimized to minimize the losses while maintaining a maximum overlap of the mode with the active region [3]. The mode still extends into the substrate, but the penetration thickness is controlled by the bottom contact layer, and the losses are much smaller due to the lack of doping in the substrate. A typical mode profile is shown in figure 1(b). This type of waveguide became subsequently known as single-metal, and, for the first few years, constituted the workhorse for the development of THz QCL devices. In the limit of a very high doping concentration (the bottom layer being replaced by a metal), the mode no longer extends into the substrate, and the resulting waveguide becomes essentially the metal-metal microstrip waveguide well known from microwave technology. Fabrication-wise, this idea was implemented for THz QCLs with wafer-bonding and subsequent removal of the original substrate. Now, with the gain medium sandwiched between two metallic layers forming a so-called double metal waveguide, one can reach confinement factors close to unity without substantially increasing the losses [10]. Though clearly superior in terms of operating temperature, double-metal THz lasers, however, display relatively lower output powers with respect to their single-metal counterparts, owing to the subwavelength mode confinement that translates in a high impedance mismatch with the outside at the waveguide end. Furthermore, beam profiles are often very divergent and irregular, mostly because of the coupling of the emitted light with the surface plasmons also existing at the external interfaces of the metallizations.

Surprisingly, though, the waveguide losses are barely increased if the top-metal is patterned, whereas the photon propagation is strongly affected. These are therefore ideal conditions for implementing novel resonator concepts, and in fact double-metal photonic crystal structures in a variety of geometries and configurations are now heavily studied as ways to engineer the laser emission properties, particularly for what concerns collimation, extraction efficiency, vertical output, etc. [11].

3 Beamshape

The applicability of THz QC lasers has till now been principally demonstrated in imaging [12] and high-resolution molecular spectroscopy [13]. Other emerging applications are in security and biomedicine or as local oscillator (LO) in a THz heterodyne spectrometer [14, 15]. Their use for plasma polarimetry and/or interferometry on the other hand is largely unexplored. Yet, available output powers routinely reach several tens of mW and can surpass the hundred in electrically pulsed operation even with relatively large duty-cycle, well suitable for propagation into relatively long and attenuating beam paths. Emission is then highly polarized, as both the selection rules of the intersubband transitions and the surface-plasmon waveguide design strongly select only TM polarization for lasing. Even if they require cooling to low temperatures, this can easily be achieved by low vibration, cryogen-free devices, like pulse tube and Sterling coolers [16, 17]. This is a crucial aspect as it allows the deployment of a self-contained compact source weighing less than 15 kg overall, with a total power consumption of no more than 240 W, requiring no further connections beyond the electrical one, and producing a reasonably Gaussian collimated beam with no external optics [17]. It can then be placed in close proximity of the plasma to be monitored, with minimal space consumption and facilitating stability and alignment issues.

The emission beam profile is a relevant property to ensure perfect coupling to the optical system, especially in interferometric measurements over relatively large distances. THz QCLs with single-metal waveguides are known to produce the most regular beam patterns, which may display a single or dual lobe nature depending on laser wavelength, device size, etc. [15]. The quality of a laser beam can be assessed by the beam-propagation factor M2, which is the ratio of the angle of divergence of a laser beam to that of an ideal Gaussian beam with the same diameter at the position of the beam waist. In ref. [16] the M2 of a 2.5 THz laser was determined by measuring the beam profile scanning a Golay cell detector with a 0.4-mm diameter aperture in a plane orthogonal to the emission direction of the QCL at different positions in front and behind the position of the beam

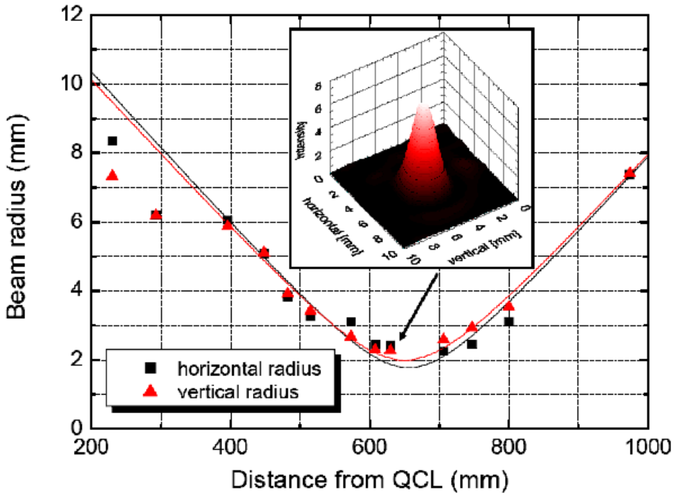

Figure 2. Propagation of the beam of a 2.5 THz QCL [16]. The beam is focused by a TPX lens ($f = 65$ mm), which is 73 mm away from the QCL. The black squares represent the horizontal beam radius while the red triangles represent the vertical radius. The black and red lines are second order polynomial fits to the measured radii. The inset shows the beam profile measured at the position of the waist.

waist created by a TPX lens. The beam diameters were determined according to the knife-edge method and according to the second moment beam width method. Both yielded the same results within the accuracy of the methods. In figure 2 the diameters of two orthogonal cuts through the beam profile are plotted as a function of distance from the QCL. The beam waist was located 650 mm from the QCL. A fit of a second order polynomial expression to the data obtained with the knife-edge method yielded a beam radius at the position of the waist of 1.8±0.1 mm and an M2 of 1.1 in the direction vertical to the layers of the superlattice of the QCL. Parallel to the layers of the superlattice the beam radius and M2 are slightly larger (2.0±0.1 mm and 1.2, respectively).

On the other side, as mentioned previously, double-metal waveguides allow the highest operating temperatures and also represent by far the best choice for frequencies below 2 THz. From an application point of view, though, such strong sub-wavelength waveguides radiate into the entire semi-sphere above the device, more similar to an antenna than to a conventional semiconductor ridge waveguide, and often present very irregular profiles. It is therefore much less efficient to collect the radiation into standard optical set-ups. Analogously to microwave technology, horn-antennas have been developed for THz QCLs, which drastically improved the directionality of the emitted power, but their integration into monolithic semiconductor devices is technically demanding [17]. For QCLs a vertical cavity design is impossible to realize, because intersubband transitions do not provide gain parallel to the growth direction. In order to still harvest the benefits of surface emission, and aiming at improving directionality and efficiency of the laser emission, research in THz QCL resonators soon turned to distributed feedback gratings, where the waveguide is corrugated with a period equal to the wavelength of the propagating mode, so that every DFB-period can be understood as a small scatterer [11]. This is again in analogy to microwave technology, where phased-array devices have a long history as directional sources. For the case of THz QCLs, the implementation of such concepts is facilitated by the fact that the optical mode

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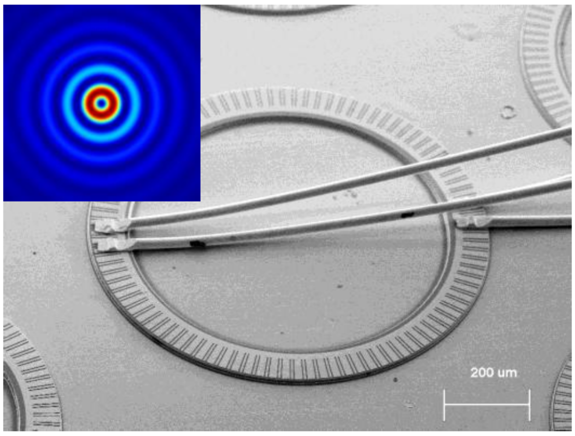

Figure 3. SEM micrograph of a 3 THz micro-ring laser. The inset shows a computed far-field of the emission, the two rings of highest intensity (red and light blue) are positioned at 3.5° and 10° with respect to the surface normal.

peaks at the metallic contact on top of the waveguide, and, therefore, is strongly influenced by any modification of the metal. The top metallization in turn can be easily modified by optical contact lithography, especially because the typical DFB-period is in the tens of microns. Due to symmetry considerations, simple second-order linear gratings are not suitable, as the vertically radiative modes present too low quality factors with respect to the in-plane emitting ones. For such reasons researchers have explored with good success alternative solutions like circular geometries, chirped two-dimensional photonic crystals, and even quasi-periodic photonic crystals [11]. While they offer excellent extraction efficiency and good beam collimation, the resulting emission profiles often deviate considerably from Gaussian, as is the case for instance of the microring devices depicted in figure 3. These, however, have been shown to provide optimal coupling to the low-loss modes of hollow metallic waveguides [19].

Finally, third order edge-emitting DFBs in metal-metal waveguides where also explored, and realized with corrugated side-walls [20]. This device geometry drastically increases directionality and radiative efficiency for longitudinal emission through matching of the propagation constant and the free space wave vector, precisely like in end-fire antenna structures. They probably represent the best compromise available right now between good spatial and spectral emission properties and excellent output and threshold performances.

4 Linewidth

In interferometric applications, the coherence properties of the laser radiation are obviously of paramount importance. In particular the coherence time (or equivalently length) is a crucial limiting factor for the allowable path differences between the interferometer arms. Naturally, for sake of compactness, stability, etc. it is preferable to minimize the path length outside the plasma reactor to be monitored, which can instead span several meters. In general the coherence time of laser light is inversely related to the emission linewidth; in semiconductor laser diodes, however, the situation is typically a bit more complex as amplitude-to-phase noise coupling often becomes the dominant source of decoherence, so that they tend to show worse properties than other laser systems. QCLs on the contrary, being based on intersubband transitions, can be ideally described by a delta-like joint density of states, and display little influence from spontaneous emission (which is highly inefficient due to the dominating non-radiative processes). As a consequence, they perform much more closely to the ideal laser noise model described originally by Schawlow and Townes; the so-called linewidth enhancement factor, which quantifies such deviation, is in fact close to zero also in THz devices [21].

As mentioned in the introduction, THz QC lasers oscillating stably on a single resonator mode can be readily achieved using the DFB concept, but also exploiting relatively short Fabry-Perot cavities, in which the free spectral range is large enough with respect to the frequency dependence of the gain to ensure a good mode selectivity. Environmental effects such as bias current and temperature variations affect the QCL frequency and are the main limiting factors of its linewidth. First measurements showed that a free running THz-QCL has a full width at half maximum (FWHM) linewidth of about 20 kHz measured within about 4 ms [15]. But for long integration times the linewidth often exceeds 10 MHz. In order to control the QCL frequency and linewidth to sub-MHz accuracy, it has to be locked to some external reference. Locking to the frequency of a THz gas laser as well as to that of a multiplied microwave source has been demonstrated [22, 23]. The achieved FWHM was several kilohertz and less than 100 Hz, respectively, and the lock condition could be maintained indefinitely. However, phase locking to a reference line from a THz gas laser is neither a very practical solution nor a very versatile technique. Frequency locking to the emission from a multiplied microwave source becomes increasingly difficult with increasing frequency due to the decrease of power from these sources. An alternative stabilization technique is based on an atomic or molecular resonance serving as frequency reference. This technique was developed shortly after the invention of the laser. Subsequently it was applied to a variety of laser types and it is now a well-established technique in most parts of the electromagnetic spectrum.

The frequency stabilization of a THz QCL relative to the center frequency of a methanol gas absorption line was reported in ref. [24]. The difference between the frequency of the absorption maximum and the QCL frequency was used to generate an error signal for the stabilization. Sub-MHz linewidth and accuracy was achieved. This approach overcomes the shortcomings of the other frequency locking schemes, because it requires only an additional detector and a small gas absorption cell while being applicable even at the highest THz frequencies due to the rich absorption spectra of molecules such as CH_3OH or H_2O. The result of the stabilization is shown in figure 4.

In the unlocked state, the derivative-like signal shows peak-to-peak variations of 15 MHz at a frequency of 1.2 Hz as well as a slow drift component. The 1.2 Hz variation is related to the temperature variation on the first cold plate of the cryocooler, which is 0.1 K as measured with a thermistor. The thermal drift is due to current heating of the QCL [25]. By monitoring the position of an absorption line as a function of time the associated frequency drift was readily observable. In the locked state the 1.2 Hz component and the thermal drift are completely eliminated. The peak-to-peak frequency variation in the locked state is approximately 600 kHz, corresponding to a long-term emission linewidth of 300±35 kHz FWHM. These values are indeed much better than the requirements posed by the plasmas dielectric constant dispersion and should translate in coherence lengths of hundred of meters, again showing the viability of QCL sources for plasma diagnostics.

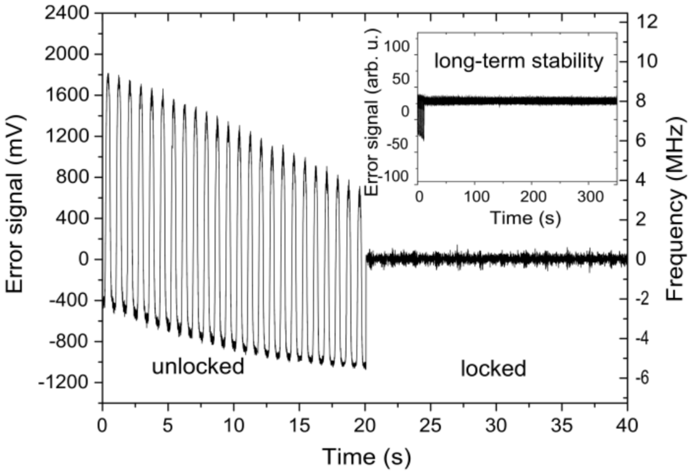

Figure 4. Error signal in the unlocked and in the locked state of the THz QCL stabilization control loop. The control loop was activated after 20 s. The variations in the unlocked state are caused by temperature and current fluctuations in the QCL. The large variation resembles the temperature cycle of the pulse tube cooler. The inset shows the long-term stability with the control loop activated after 12 s.

5 Tunability and modulation

Tuning the emission wavelength of QCLs operating in the terahertz region is notoriously difficult, but has great importance for lasers in applications such as spectroscopy and sensing. In single-mode DFB lasers for instance, the grating period is “written” into the semiconductor laser crystal that, in turn, has low elasticity: tuning the grating periodicity — and thus the emission frequency — mechanically is then really difficult. Therefore, the frequency tuning of a single-mode DFB laser typically relies on changing the effective refractive index by varying the heat sink temperature, or the applied electric current/voltage. This means that, although DFB-QCLs are very useful spectroscopic sources thanks to their small size, predictable single-mode emission and mode-hop free tuning capability, the achievable tuning range is actually limited to a few GHz.

A detailed evaluation of the tuning rate of THz QCLs is conventionally performed via heterodyne experiments employing Schottky diode mixer detectors. The thermal frequency tuning depends on the specific device geometry, on the drive current and temperature range, as well as on the specific lasing mode. To date, tuning rates varying from -34 MHz/K to almost -100 MHz/K have been reported [26]. On the other hand, for some active region designs, positive tuning rates with current are also observed up to $\sim$ 8 MHz/mA [26]. This effect, opposite to the temperature one, originates from a mode-pulling effect, which is produced by the blue-shift of the underlying gain profile with applied bias, with the ensuing shift of the resonant contribution to the material refractive index (see figure 5).

Spectroscopy of solid features in the far-infrared typically requires a tunability range well above 100 GHz. To address the latter requirements a different approach based on external cavity (EC) lasers with frequency selective feedback can be used. In the latter case, a tunable feedback is provided by the insertion of a controllable optical element such as a diffraction grating or a simple movable mirror. However, the implementation of conventional external cavity concepts is quite

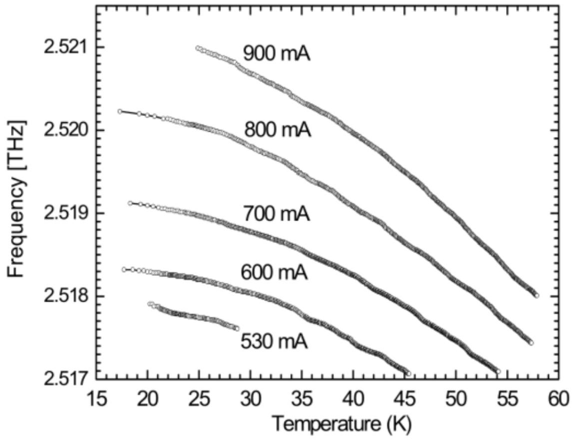

Figure 5. Emission frequency of a 2.5 THz DFB QCL as a function of temperature and current [26].

hard in the case of THz semiconductor lasers, since the waveguide cross-sections are considerably smaller than the free-space wavelength. It is therefore challenging to efficiently couple the radiation to an external element and back into the waveguide. Few successful reports of tunable external cavity THz QCLs have been reported in the last few years, by employing different optical approaches [27]. Continuous tuning was anyway limited to max 2% of the emission frequency.

Micro-mechanical tuning solutions on the emission wavelength scale of THz QCLs can instead be implemented. In a laser resonator, the greatest relative change of mode size and/or overlap can indeed be obtained when the length scale of the varied cavity dimension is of the order of the wavelength or possibly even below. Since in this case the frequency dispersion per unit displacement is proportional to ω/λ, a mechanical tuning on a length scale coincident with the emission wavelength of THz QCLs (from several tens of μm to a few hundred μm) is enough to achieve huge frequency variations. Exploiting vertical emission from second-order double-metal distributed feedback lasers, it is for instance possible to realize an external cavity arrangement in which, instead of varying the frequency that is fed back into the laser, one controls the detuning of an external cavity of size comparable to the radiation wavelength with respect to the lasing mode of the strongly coupled laser resonator. Such a system can be simply realized by placing a piezo-actuated metallic surface at wavelength distance above the DFB grating, so that the metallic grating acts as a coupler between the laser waveguide and the external one forming between grating and metal. By changing the position of the metallic element, the energy of the modes of the external waveguide moves across that of the lasing DFB mode, and the laser emission is then tuned by approximately twice the mode anti-crossing frequency. In this preliminary experiment a maximum tuning range of 20 GHz has been obtained [27]. More recently, single mode continuous frequency tuning via an opto-mechanical approach showed great promise for a broad tuning covering the entire laser gain spectrum. The technique 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 ($w \approx \lambda/3$), 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

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the grating, the tuning is obtained by changing the transverse wavevector by mechanically bringing another material (plunger) close to the flat side of the laser ridge, thus influencing the mode. The use of narrow ridges is indeed beneficial to increase the QCL tunability range, since the fraction of the optical mode propagating outside the waveguide is further extended. In addition, MEMS-based plungers overcome the limitations induced by the large friction between the plunger and its movement track as well as by the lack of a restoring mechanism. With this elegant technical solution a continuous and reversible tuning range of 330 GHz in single mode has been obtained [28].

What is perhaps more relevant though to applications in plasma diagnostics is the possibility of directly modulating the laser output at very high speed. As with conventional laser diodes, this is achieved by directly modulating the drive current, but, owing to the unipolar nature of QC devices, the modulation frequency is not limited by relaxation oscillations, unlike in interband devices. For this reason frequencies even above 10 GHz can be achieved without too much effort by simply taking care of the appropriate high frequency design of mounting and packaging [29]. These values are clearly way above the necessities of plasma monitoring, where modulation can be mostly implemented to improve the signal-to-noise and eliminate contributions arising from low frequency vibrations, etc. It would also ensure the possibility to detect changes of the density and magnetic field even on very fast timescales. It has to be noted that direct modulation of the laser drive current produces both an AM and an FM modulation of the laser output. This would probably open the way to new possibilities and techniques in the diagnostics, borrowing from the concepts developed for high-sensitivity laser diode spectroscopy.

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