

1.3.17 Quantum engineering of THz quantum cascade lasers and applications to quantum metrology

Quantum cascade lasers (QCLs) represent a fascinating accomplishment of quantum engineering and enable the direct generation of terahertz (THz) frequency radiation from an electrically-biased semiconductor heterostructure. Their large spectral bandwidth, high output powers and quantum-limited linewidths have facilitated the realization of THz pulses by active mode-locking and passive generation of optical frequency combs (FCs) through intracavity four-wave-mixing. Here we present recent results in the implementation of THz QCLs into advanced photonic structures and report on state-of-the-art applications in quantum-metrology and high-resolution spectroscopy.

The development of modern nanofabrication technologies, combined with new laser resonator concepts, has recently enabled the control and confinement of electron and photon paths, in optoelectronic devices, with an unprecedented degree of control. For example, microcavities, photonic crystals, and both pseudo-random and random photonic structures, can manipulate and confine light in small volumes, and at targeted frequencies. This has further expanded the functionality of the THz QCL, allowing operation at a single emission frequency or over a broad (0.5 THz) frequency bandwidth, with designed directional beam patterns.

We demonstrated double-metal waveguided distributed feedback THz quantum cascade wire lasers, exploiting an innovative approach in which feedback is provided by a lateral sinusoidal ridge corrugation, while light extraction is separately controlled by a hole array in the top metallization. In this case, the periodicity of the array of surface holes is not an integer multiple of the lateral corrugation controlling the feedback. The feedback grating selects the lasing frequencies and allows robust single mode emission, exploiting the inherently high spectral purity of THz QCLs, while the extraction array is finely tuned to optimize the radiation outcoupling (Fig.1a). This new architecture, extendable to a broad range of far-infrared frequencies, has led to the achievement of low-divergent beams (10°), single-mode emission, high slope efficiencies (250 mW/A), and stable CW operation.

By exploiting a broadband heterostructure, we furthermore developed the first one-dimensional quasi-crystal distributed feedback laser by lithographically patterning a series of air slits of different widths, following the Octonacci sequence, on the top metal layer of a THz QCL. We tuned the emission from single-mode to multi-mode over a 530 GHz bandwidth, achieving a maximum peak optical power of 240 mW (190 mW) in multimode (single-mode) lasers with record slope efficiencies up to ≈ 570 mW/A at 78 K and ≈ 700 mW/A at 20 K, wall-plug efficiencies of $\eta \approx 1\%$ and low divergent emission (Fig. 1b).

To push the concept of disordered resonators further, we also developed the first electrically pumped CW random laser (Fig. 1c). By combining this concept with the QCL gain media we obtain a highly collimated vertical emission at ~ 3 THz, with a 430 GHz bandwidth, device operation up to 110 K, peak (pulsed) power of 21 mW, CW emission of 1.7 mW and continuous frequency tuning over 11 GHz.

The same broadband heterostructure can be also exploited to develop miniaturized frequency comb (FC) sources across hard-to-access spectral regions, as the far-infrared. Four-wave-mixing based QCL-combs are ideal candidates, in this respect, due to the unique possibility to tailor their spectral emission by proper nanoscale design of the quantum wells. We demonstrate full-phase-

stabilization of record dynamic range, high power QCL-comb against the primary frequency standard, proving independent and simultaneous control of the two comb degrees of freedom (modes spacing and frequency offset) at a metrological level (Fig.1d). Each emitted mode exhibits sub-Hz relative frequency stability, while a correlation analysis on the modal phases confirms the high degree of coherence in the device emission, proving that this technology is mature for metrological-grade uses, as well as for an increasing number of scientific and technological applications.

Also, by integrating an on-chip tightly coupled mirror with the QCL cavity (Gires Tournois interferometer) we demonstrate tunable, lithographically independent, control of the free-running coherence properties of THz QCL FCs.

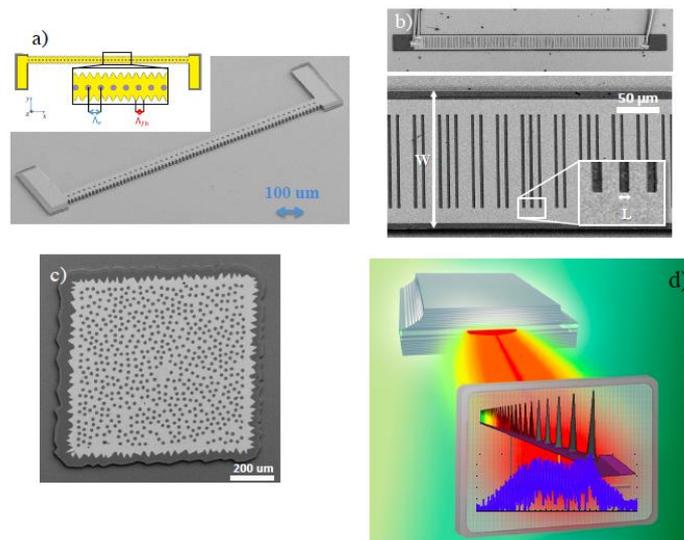


Figure 1. (a) Schematic diagram of the laterally corrugated wire laser and related SEM picture; (b) SEM image of an Octonacci quasi crystal THz laser; (c) SEM image of a random 2D THz QCL resonator; (d) Quantum cascade laser emitting a discrete set of equally spaced modes, each corresponding to the tooth of an optical frequency comb. The semiconductor heterostructure embedded in the QCL active core is schematically shown as a sequence of striped lines.

Tunable oscillators are a key component of almost all electronic and photonic systems. Yet, a technology capable of operating in the terahertz (THz)-frequency range and fully suitable for widescale implementation is still lacking. This issue is significantly limiting potential THz applications in gas sensing, high-resolution spectroscopy, hyper-spectral imaging and optical communications. The THz QCL is arguably the most promising solution in terms of output power and spectral purity. In order to achieve reliable, repeatable and broad tuneability, we exploited the strong coupling between two different cavity mode concepts: a distributed feedback one-dimensional photonic resonator (providing gain), and a mechanically actuated wavelength-size microcavity (providing tuning). The result is a continuously tunable, single-mode emitter covering a 162 GHz spectral range, centered on 3.2 THz with a few tens of MHz resolution and an unprecedented compact and simple architecture (Fig. 2a-b).

In parallel, we also developed a novel approach to couple THz radiation from a double-metal QCL into an on-chip hollow rectangular waveguide feeding a triangular horn, with the specific aim of optimizing the optical beam divergence. The conceived novel extractor is composed of three parts: a series of slits

patterned at the end of the laser top contact (slit coupler), a metallic waveguide section (feeder) assembled on top of the laser itself, and an adiabatic expansion of the feeder, forming a horn that radiates into an optical fiber or, alternatively, into free space (Fig. 2c-e). The developed waveguide component is capable of efficiently shaping the output beam profile from a double-metal THz QCL simultaneously reducing the facet reflectivity opening the way to the realization of more complex systems such as QCL amplifiers or external cavity tuners for multi-spectroscopy applications, injection seeding and metrological approaches across the far-infrared.

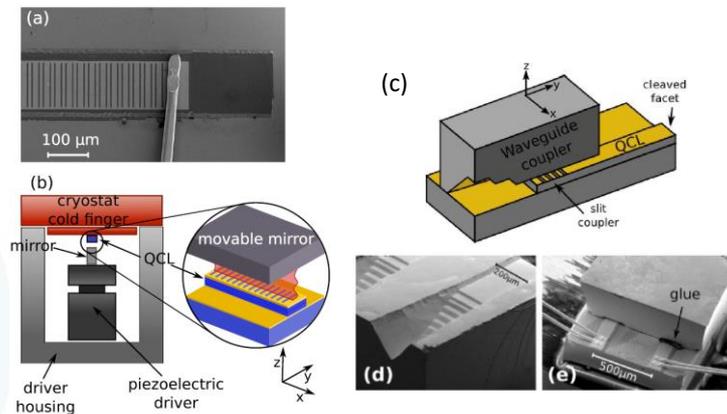


Figure 2. (a) Scanning electron microscope (SEM) image of a dual-slit DFB QCL. The absorbing boundary is visible around the grating. (b) Schematic diagram of the external cavity arrangement. The movable mirror was milled from an aluminum block and was then laid on top of the piezoelectric actuator. (c) Device schematics: yellow areas indicate metallized surfaces, gray corresponds to GaAs (d) SEM image of the cleaved chip containing two waveguide couplers; (e) SEM image of the final assembly, taken from the cleaved facet side.

We also pioneered a novel approach to optimize extraction efficiency and collimation of the output radiation of single-mode THz QCLs by developing quasi-crystalline resonators in which the distinction between symmetric (vertically radiative, but low-quality factor, Q) and antisymmetric (non-radiative, high- Q) modes is fully overcome, therefore elegantly circumventing any power cancellation issue in the far-field. Our 2D photonic quasi-crystal THz QCLs based on a Penrose P2 (kite and dart) tiling with a five-fold rotational symmetry, reached 0.1-0.2% wall-plug efficiencies and 65mW peak output powers with characteristic surface-emitting conical beam profiles, result of the rich quasi-crystal Fourier spectrum (Fig. 3a-b).

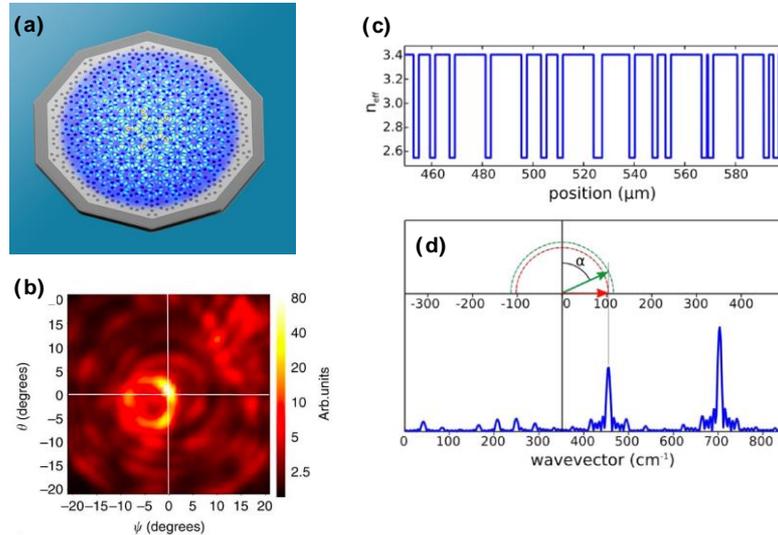


Figure 3. (a) SEM image of a prototype quasi-crystal resonator device. The lattice spatial length scale a was lithographically designed at each vertex of a Penrose pattern and imprinted into the top Cr/Au metallization of the THz QCL (see inset). The computed 2D spatial profiles of the electric field modulus for the higher quality factor optical mode is shown on the top of the SEM picture (b) Far-field measured from the device with $a=21 \mu\text{m}$; (c) Spatial dependence of the effective refractive index of a bi-periodic DFB THz QCL. (d) Spatial spectra of the grating of panel (c). Two light cones (dashed lines) are drawn, corresponding to the two bandgap modes of a first order grating with spatial periodicity of 712 cm^{-1} . The two modes are predicted to be at 3.1 THz (red, inner circle) and 3.35 THz (green, outer circle), and to radiate at different angles, as indicated by the arrows.

We then worked on photonic engineering solutions to control the emission frequency and the output beam direction of one-dimensional DFB THz QCLs, independently. The spatial refractive index modulation of the gratings necessary to provide optical feedback at a fixed frequency and, simultaneously, a far-field emission pattern centered at controlled angles, was designed through use of an appropriate wavevector scattering model (Fig. 3c-d). Single-mode THz emission at angles tuned by design between 0° and 50° was realized, leading to an original phase-matching approach, lithographically independent, for highly collimated THz QCLs.

We then developed intriguing application systems based on the described THz QCL resonators.

We demonstrate real-time digital holography (DH) at THz frequencies exploiting the high spectral purity and the mW output power of a QCL combined with the high sensitivity and resolution of a microbolometric array. We show that, in a one-shot exposure, phase and amplitude information of whole samples, either in reflection or in transmission, can be recorded. Furthermore, a 200 times reduced sensitivity to mechanical vibrations and a significantly enlarged field of view are observed, as compared to DH in the visible range. These properties of THz DH enable unprecedented holographic recording of real-world dynamic scenes.

We demonstrate a metrological-grade THz spectroscopy system based on the combination of a free space THz frequency-comb synthesizer (FCS) and a THz QCL. The QCL is phase locked to the free-space THz FCS, and its frequency is swept across a methanol transition by tuning the comb repetition rate, which is ultimately disciplined by the Cs primary frequency standard. The absolute frequency scale provides an uncertainty of a few parts in 10^{-11} on the laser

frequency and 10^{-9} on the line-center determination, ranking this technique among the most precise ever developed in the THz range.

References

- [1] L. Consolino, M. Nafa, F. Cappelli, K. Garrasi, F.P. Mezzapesa, L. Li, A.G. Davies, E.H. Linfield, M.S. Vitiello, P. De Natale, S. Bartalini, *Fully phase-stabilized quantum cascade laser frequency comb*, Nature Communications **10**, 2938 (2019).
- [2] S. Biasco, H.E. Beere, D.A. Ritchie, L. Li, A.G. Davies, E.H. Linfield, M.S. Vitiello, *Frequency-tunable continuous-wave random lasers at terahertz frequencies*, Nature Light: Science & Applications **8** (1) (2019).
- [3] F.P. Mezzapesa, V. Pistore, K. Garrasi, L. Li, A.G. Davies, E.H. Linfield, S. Dhillon, M.S. Vitiello, *Tunable and compact dispersion compensation of broadband THz quantum cascade laser frequency combs*, Optics Express **27** (15), 20231 (2019).
- [4] L. Consolino, A. Campa, D. Mazzotti, M.S. Vitiello, P. De Natale, S. Bartalini, *Bow-tie cavity for terahertz radiation*, Photonics **6** (1) (2019).
- [5] K. Garrasi, F.P. Mezzapesa, L. Salemi, L. Li, L. Consolino, S. Bartalini, P. De Natale, A.G. Davies, E.H. Linfield, M.S. Vitiello, *High dynamic range, heterogeneous, terahertz quantum cascade lasers featuring thermally tunable frequency comb operation over a broad current range*, ACS Photonics **6**, 73 (2019).
- [6] L.H. Li, K. Garrasi, I. Kundu, Y.J. Han, M. Salih, M.S. Vitiello, A.G. Davies, E.H. Linfield, *Broadband heterogeneous terahertz frequency quantum cascade laser*, Electronics Letters **54**, 1229 (2018).
- [7] S. Biasco, K. Garrasi, F. Castellano, L. Li, H.E. Beere, D.A. Ritchie, E.H. Linfield, A.G. Davies, M.S. Vitiello, *Continuous-wave highly-efficient low-divergence terahertz wire lasers*, Nature Communications **9**, 1 (2018).
- [8] L. Consolino, S. Jung, A. Campa, M. De Regis, S. Pal, J.H. Kim, K. Fujita, A. Ito, M. Hitaka, S. Bartalini, P. De Natale, M.A. Belkin, M.S. Vitiello, *Spectral purity and tunability of terahertz quantum cascade laser sources based on intracavity difference-frequency generation*, Science Advances **3**, e1603317 (2017).
- [9] S.S. Dhillon, M.S. Vitiello et al., *The 2017 terahertz science and technology roadmap*, Journal of Physics D: Applied Physics **50** (4), 043001 (2017).
- [10] S. Biasco, L. Li, E.H. Linfield, A.G. Davies, M.S. Vitiello, *Multimode, aperiodic terahertz surface-emitting laser resonators*, Photonics **3** (2), 32 (2016).
- [11] A. Sampaolo, P. Patimisco, M. Giglio, M.S. Vitiello, H.E. Beere, D.A. Ritchie, G. Scamarcio, F.K. Tittel, V. Spagnolo, *Improved tuning fork for terahertz quartz-enhanced photoacoustic spectroscopy*, Sensors **16**, 439 (2016).
- [12] F. Castellano, L. Li, E.H. Linfield, A.G. Davies, M.S. Vitiello, *Frequency and amplitude modulation of ultra-compact terahertz quantum cascade lasers using an integrated avalanche diode oscillator*, Scientific Reports **6**, 23053 (2016).
- [13] R. Degl'Innocenti, Y.D. Shah, L. Masini, A. Ronzani, A. Pitanti, Y. Ren, D.S. Jessop, A. Tredicucci, H.E. Beere, D.A. Ritchie, *Hyperuniform disordered terahertz quantum cascade laser*, Scientific Reports **6**, 193225 (2016).
- [14] F.P. Mezzapesa, L.L. Columbo, C. Rizza, M. Brambilla, A. Ciattoni, M. Dabbicco, M.S. Vitiello, G. Scamarcio, *Photo-generated metamaterials induce modulation of CW terahertz quantum cascade lasers*, Scientific Reports **5**, 16207 (2015).
- [15] F. Castellano, V. Bianchi, L. Li, J. Zhu, A. Tredicucci, E.H. Linfield, A.G. Davies, M.S. Vitiello, *Tuning a microcavity-coupled terahertz laser*, Applied Physics Letters **107**, 261108 (2015).
- [16] M. Locatelli, M. Ravaro, S. Bartalini, L. Consolino, M.S. Vitiello, R. Cicchi, F. Pavone, P. De Natale, *Real-time terahertz digital holography with a quantum cascade laser*, Scientific Reports **5**, 13566 (2015).
- [17] V. Spagnolo, P. Patimisco, R. Pennetta, A. Sampaolo, G. Scamarcio, M.S. Vitiello, F.K. Tittel, *THz Quartz-enhanced photoacoustic sensor for H₂S trace gas detection*, Optics Express **23**, 7574 (2015).
- [18] F. Castellano, L. Li, E.H. Linfield, A.G. Davies, H.E. Beere, D.A. Ritchie, M.S. Vitiello, *THz waveguide adapters for efficient radiation out-coupling from double metal THz QCLs*, Optics Express **23**, 5190 (2015).

- [19] M.S. Vitiello, G. Scalari, B. Williams, P. De Natale, *Quantum cascade lasers: 20 years of challenges*, Optics Express **23** (4), 5167 (2015).
- [20] A. Campa, L. Consolino, M. Ravaro, D. Mazzotti, M.S. Vitiello, S. Bartalini, P. De Natale, *High-Q resonant cavities for terahertz quantum cascade lasers*, Optics Express **23**, 3751 (2015).
- [21] L. Consolino, A. Campa, M. Ravaro, D. Mazzotti, M.S. Vitiello, S. Bartalini, P. De Natale, *Saturated absorption in a rotational molecular transition at 2.5 THz using a quantum cascade laser*, Applied Physics Letters **106**, 021108 (2015).
- [22] F. Castellano, S. Zanotto, L.H. Li, A. Pitanti, A. Tredicucci, E.H. Linfield, A.G. Davies, M.S. Vitiello, *Distributed feedback terahertz frequency quantum cascade lasers with dual periodicity gratings*, Applied Physics Letters **106**, 011103 (2015).
- [23] M.S. Vitiello, M. Nobile, A. Ronzani, A. Tredicucci, F. Castellano, V. Talora, L. Li, E.H. Linfield, A.G. Davies, *Photonic quasi-crystal terahertz lasers*, Nature Communications **5** (1), 1 (2014).
- [24] F. Mezzapesa, M. Petruzzella, M. Dabbicco, H. Beere, D. Ritchie, M.S. Vitiello, G. Scamarcio, *Continuous-wave reflection imaging using optical feedback interferometry in terahertz and mid-infrared quantum cascade lasers*, IEEE Transactions on Terahertz Science and Technology **4**, 631 (2014).
- [25] S. Bartalini, L. Consolino, P. Cancio, P. De Natale, P. Bartolini, A. Taschin, M. De Pas, H. Beere, D. Ritchie, M.S. Vitiello, R. Torre, *Frequency-comb-assisted terahertz quantum cascade laser spectroscopy*, Physical Review X **4**, 021006 (2014).
- [26] F.P. Mezzapesa, L.L. Columbo, M. Brambilla, M. Dabbicco, M.S. Vitiello, G. Scamarcio, *Imaging of free carriers in semiconductors via optical feedback in terahertz quantum cascade lasers*, Applied Physics Letters **104**, 041112 (2014).
- [27] P. Patimisco, S. Borri, A. Sampaolo, H.E. Beere, D.A. Ritchie, M.S. Vitiello, G. Scamarcio, V. Spagnolo, *A quartz enhanced photo-acoustic gas sensor based on a custom tuning fork and a terahertz quantum cascade laser*, Analyst **139**, 2079 (2014).