



# Sculpting harmonic comb states in terahertz quantum cascade lasers by controlled engineering

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Optical frequency combs (OFCs), which establish a rigid phase-coherent link between the microwave and optical domains of the electromagnetic spectrum, are emerging as key high-precision tools for the development of quantum technology platforms. These include potential applications for communication, computation, information, sensing, and metrology and can extend from the near-infrared with micro-resonator combs, up to the technologically attractive terahertz (THz) frequency range, with powerful and miniaturized quantum cascade laser (QCL) FCs. The recently discovered ability of the QCLs to produce a harmonic frequency comb (HFC)—a FC with large intermodal spacings—has attracted new interest in these devices for both applications and fundamental physics, particularly for the generation of THz tones of high spectral purity for high data rate wireless communication networks, for radio frequency arbitrary waveform synthesis, and for the development of quantum key distributions. The controlled generation of harmonic states of a specific order remains, however, elusive in THz QCLs. Here, and by design, we devise a strategy to obtain broadband HFC emission of a pre-defined order in a QCL. By patterning  $n$  regularly spaced defects on the top surface of a double-metal Fabry–Perot QCL, we demonstrate harmonic comb emission with modes spaced by an  $(n + 1)$  free spectral range and with an optical power/mode of  $\sim 270 \mu\text{W}$ . © 2024 Optica Publishing Group under the terms of the Optica Open Access Publishing Agreement

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## 1. INTRODUCTION

Quantum cascade lasers (QCLs) [1] emitting at terahertz frequencies (1.2–5.7 THz) [2,3] are unipolar semiconductor lasers characterized by ultrafast gain recovery dynamics ( $\sim 20$  ps) [4] and a large third-order  $\chi^{(3)}$  Kerr nonlinearity [5,6]. These features, combined with their small footprint, intrinsic spectral purity [7], continuous wave (CW) operation, and relatively high output powers ( $> 2$  W) [8], make these sources an ideal platform for the observation of mode-locked multimode regimes [9] and quantum correlation effects [7]. The experimental discovery that QCLs can spontaneously emit optical frequency combs (OFCs) [9–11] has dramatically increased their technological potential in metrology [12], communication [13], sensing [14], dual-comb imaging and spectroscopy [15,16], and quantum technologies [14,17,18].

OFCs operating in the harmonic regime [harmonic frequency combs (HFCs)] represent a promising technology [19,20]. In contrast to the standard, evenly spaced comb operation, where the mode separation matches the cavity round trip frequency, in the harmonic state, optical power is concentrated in a few modes spaced by several multiples of the cavity free spectral range (FSR) [21]. For both QCL FCs and HFCs, quantum correlations are

expected to be established by resonant four-wave-mixing (FWM) [9,14,22,23] enabled by the third-order  $\chi^{(3)}$  Kerr nonlinearity, which is the mechanism responsible for QCL comb mode proliferation and for frequency- and phase-locking the side modes [5,6]. FWM has also been identified as the core mechanism of entanglement among laser modes in passive micro-resonator-based comb emitters [24], triggering an increased interest for the exploitation of the same effect in standard Fabry–Perot (FP) QCL FCs.

Under pure electrical injection, the frequency spacing of the THz QCL combs is fixed by the length of the cavity and by the effective group index of the waveguide [25]. The two types of coherent regime, FC and HFC, can, however, co-exist in the same device for different values of the DC current, as has been observed in both mid-infrared (mid-IR) [19,26,27] and THz [28] QCLs. In general, coherent regimes alternate with irregular unlocked states upon sweeping the bias current [29]. However, to date, experimental access to HFC states in electrically driven THz QCLs has been limited. The only mechanism to control these devices is through the injected current. Usually, the QCL first reaches a state of high single-mode intracavity intensity, and when this intensity is large

enough, an instability threshold caused by the  $\chi^{(3)}$  population pulsation nonlinearity is reached, favoring the appearance of modes separated by tens of the free spectral range (FSR) from the first lasing mode [28].

Controlled generation of harmonic states of a specific order has been achieved in mid-IR QCLs either by the injection of an optical seed into the laser cavity [30], or through defect engineering in mid-IR ring QCLs [31]. However, similar attempts remain elusive in THz QCLs. To date, active harmonic mode-locking in THz QCLs has only been realized by injection locking through an active modulation and via direct translation to harmonic pulse generation using an ultrafast setup [32]. Although the latter approach is effective for long (3–6 mm) devices and low harmonic orders, it is not a passive approach, it requires additional equipment for generating the microwave current, and it becomes impractical for short QCLs and higher-order harmonics.

Here, we propose an experimental methodology that alleviates the influence of device fabrication and operation uncertainties on the emission state of the FC, allowing the specific order of the HFC to be determined in advance. We engineer, by design, broadband HFCs with a tailored number of harmonic modes of a pre-defined order in standard FP cavity THz QCLs. By employing a heterogeneous gain medium [33,34], and a cavity architecture that integrates a fixed number of  $n$  scattering points (defects) placed at designed points along the top surface of the double-metal QCL cavity, we demonstrate harmonic comb emission over the entire operational range of the QCL, with harmonic modes spaced by  $(n + 1)$  FSR. Our approach adopts a standard linear cavity, rather than a ring cavity, and does not require external modulation which would necessitate complex external circuitry. The ability to control HFC generation by design in THz QCLs paves the way for novel applications, such as the generation of multi-mode squeezed states of light, arising from quantum-correlated side-band modes.

## 2. SAMPLE DESIGN

Figure 1(a) shows the schematic diagram of a prototypical device, comprising an edge-emitting double-metal waveguide QCL of length  $L$  with  $n = 2$  defects patterned on the top contact in the form of narrow rectangular slits covered by a thin layer (5 nm) of nickel.

The equidistant defects divide the cavity into  $n + 1$  parts spaced by  $L/(n + 1)$ , creating a periodic pattern on the top contact. This affects the spatial dependence of the intermode beat intensity along the cavity, originating from the interference of any combination of frequency comb modes.

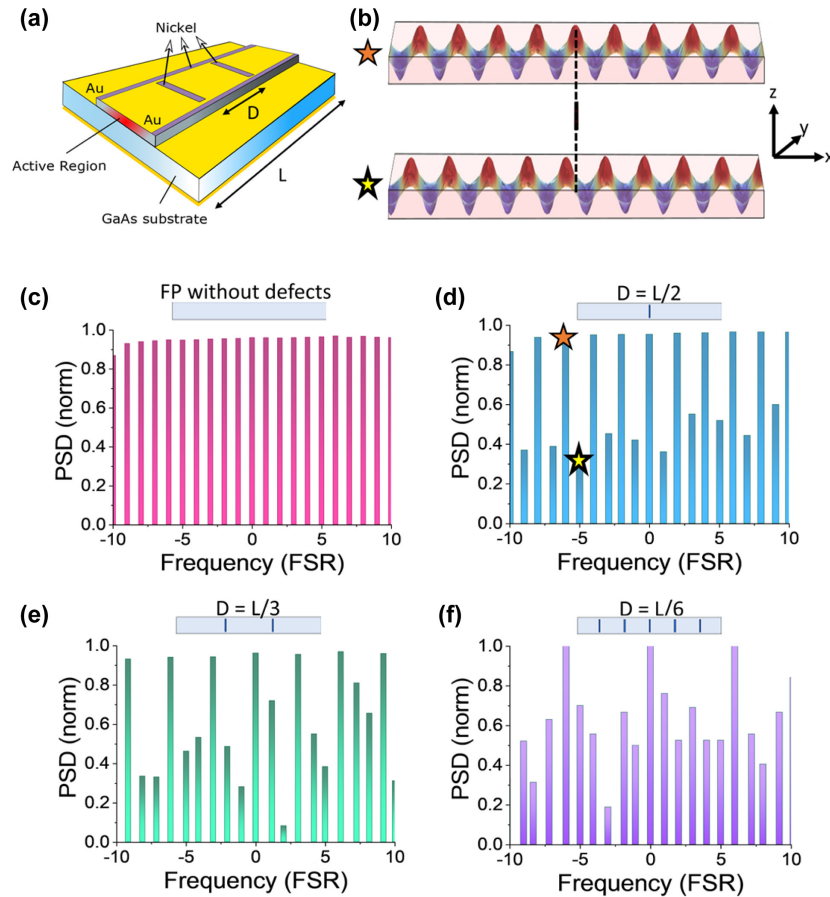
The optical modes sustained by the resonator are determined by  $L$ , as in any FP structure, but the sequence of patterned defects acts as a frequency filter that introduces periodic additional losses. This leads to an intensity modulation of the frequency of the modes supported by the cavity. We perform three-dimensional simulations via a finite element method (Comsol Multiphysics) to evaluate the effect induced by the surface patterning on the  $TM_{00}$  QCL modes. Details of the simulation parameters are provided in the Supplement 1. The simulations show that the losses induced by the defects strongly depend on the defects' spatial position with respect to the field phase. Figure 1(b) displays the electric field distribution inside the QCL cavity with one defect placed in the center of the cavity ( $L/2$ ). This design is intended for the generation of a second harmonic FC. The optical modes peak at the metallic cladding on

the top of the active region. Two prototypical neighboring modes of the comb with frequencies of 3.009 THz and 2.992 THz are shown in the top and bottom panels of Fig. 1(b), respectively. They correspond to a harmonic and a non-harmonic mode, respectively. The black rectangle and dashed line between the panels indicate the defect position on the top contact of the cavity. In the first case (top), the defect position is aligned with a peak of the electric field, while in the second case (bottom), the defect and the field peaks are misaligned. The effects of the defects on the electric field distribution in the in-plane and out-of-plane directions are numerically computed (Comsol Multiphysics) for two prototypical devices (see Supplement 1).

We can evaluate the effect induced by the defects on the electric field by looking at the redistribution of the field's components. Due to the intersubband nature of the device, the photons propagating in the cavity are transverse-magnetic (TM) polarized, i.e., with the electric field parallel to the growth direction ( $z$ ). The defects redistribute the electric field components, thereby reducing the  $E_z$  field amplitude and introducing additional losses to the modes whose electric field maximum/minimum is not aligned with the defect (see Supplement 1).

To quantify the field redistribution introduced by the defects, we can extract the ratio  $E_z/|E_{\text{tot}}|$ , where  $\vec{E}_{\text{tot}} = E_x + E_y + E_z$  is integrated over the entire cavity. The comparison between the reference structure without defects shows that  $E_z/|E_{\text{tot}}|$  drops, on average, by 0.7% when the defects are all aligned with the field peaks, while it drops by 11% for the frequencies for which the defects and field peaks are misaligned, i.e., the non-harmonic modes. This field redistribution resembles a mode-dependent radiative loss modulation. We calculate the radiative losses through the 3D quality factor ( $Q$  factor), extrapolated as a direct output of the simulations (see Supplement 1). If we express the harmonic modulation as the ratio of the average radiative losses between the harmonic and non-harmonic modes, then we find that by introducing a defect in the center of the cavity, the radiative losses are almost the same for the harmonic modes, but they are 20% higher for the non-harmonic ones. Finally, upon introducing a lossy material, e.g., a thin nickel layer covering the defect, the losses increase for all modes, particularly for the non-harmonic ones, leading to an increase of the harmonic modulation of 22.3% (see Supplement 1).

We next simulate the spectral response of the defects on the cavity modes in a range of 20 FSR around 3 THz, i.e., the central frequency of the QCL employed here [33,34]. Figures 1(c)–1(f) show the normalized power spectral density (PSD) calculated in the presence of an increasing number of defects. The corresponding geometry is illustrated in the insets at the top of each panel. Figure 1(c) shows the results obtained in a standard FP cavity without defects and acts here as our reference. In Figs. 1(d)–1(f), we show the simulated results for a waveguide divided by equidistant defects, with  $n = 1$  [Fig. 1(d)], 2 [Fig. 1(e)], and 5 [Fig. 1(f)]. In the defect-free FP cavity (reference), the simulated PSD reveals a series of equidistant modes of comparable intensity, as expected. Conversely, when the defects are implemented, the PSD presents a visible frequency-dependent modulation that affects all comb modes except those at the  $n^{\text{th}}$  harmonic.



**Fig. 1.** Device concepts and simulated frequency dependence of the power spectral density. (a) Three-dimensional device structure. The top contact of a double-metal QCL waveguide is patterned with a periodic number of slits covered by a 5-nm-nickel layer. To obtain operation on the  $n$ -harmonic state, the distance between the slits is set to  $L/(n + 1)$ . (b) Schematic diagram of the electric field distribution inside a QCL cavity with one defect placed at  $L/2$ . The upper and lower panels represent two neighboring modes of the comb. The black rectangle and dashed line indicate the position of the defect on the top contact of the cavity. Changing the frequency also changes the relative position of the field peaks with respect to the defect. (c)–(f) Simulated field power spectral density (PSD) of the field for different cases. A FP cavity without defects (c) shows a PSD equally distributed between all the modes. In the presence of defects (d)–(f), the frequencies for which the field peaks are not aligned with the defects show more significant losses resulting in a spectrum with a harmonic modulation. All spectra are centered with respect to 3 THz, and the frequency axis is normalized by the cavity FSR. The insets show the number and distribution of defects on the top contact of the cavity. The orange and yellow stars indicate, respectively, the case in which the defect is aligned with the field peaks and the one where it is misaligned (b).

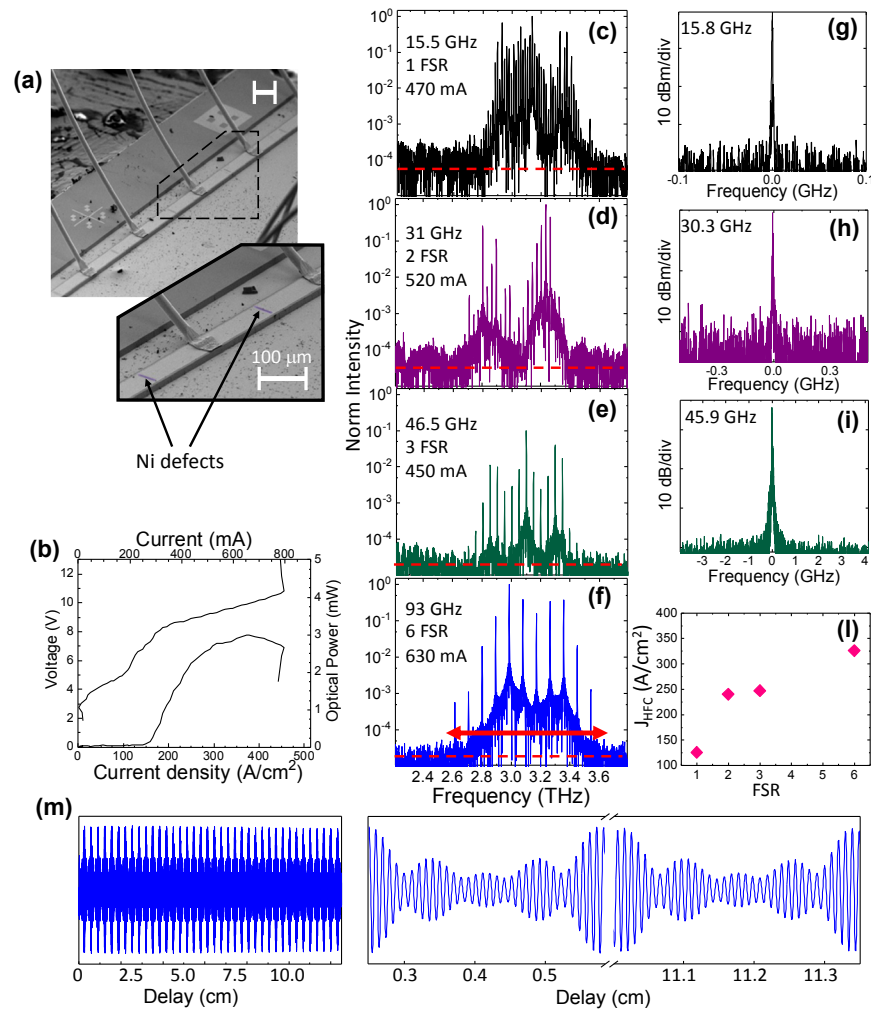
### 3. SAMPLE FABRICATION AND CHARACTERIZATION

A set of QCLs were then fabricated with the number of defects  $n$  varying in the range 0–5. We employed a 17- $\mu\text{m}$ -thick GaAs/AlGaAs heterostructure with an active region comprising three active modules, each tuned to a different central frequency, and each based on longitudinal optical phonon-assisted interminiband transitions. All stacks show comparable threshold current densities ( $J_{\text{th}}$ ) [33] and gain bandwidths centered at 2.5 THz, 3 THz, and 3.5 THz (see Section 2). The choice of a heterogeneous heterostructure guarantees the design of a harmonic spectral response broader than those achieved so far using only bias control in a single-frequency laser [28]. The double-metal FP laser cavity includes top Ni-based lossy side absorbers to inhibit high-order lateral modes by increasing their threshold gains [35] (Appendix A). The defects were lithographically defined onto the top Au layer, exposing the active region below and then covered with a 5-nm-thick Ni layer [Fig. 2(a)]. For consistency, all QCLs are fabricated with an equal width of 70  $\mu\text{m}$  and a length of  $L = 2.5$  mm.

The current–voltage (I–V) and light–current (L–I) characteristics of a prototypical QCL with  $n = 5$  is reported in Fig. 2(b). The device has a threshold current density of  $J_{\text{th}} = 165 \text{ A/cm}^2$ , 15% higher than that of the reference structure [10] and shows a consistent dynamic range  $J_{\text{max}}/J_{\text{th}} = 3.5$ , a maximum optical power of 3 mW, a slope efficiency of 16.5 mW/A and a 0.05% wall-plug (WP) efficiency. A comparison of the main figures of merit of the complete set of fabricated devices is provided in the Supplement 1. Compared to the reference FC QCL, the HFCs show a 4%–20% increase in the threshold current density ( $J_{\text{th}}$ ), a corresponding 5%–35% increase of the slope efficiency, a 20%–29% variation of the WP efficiency, and a 16%–22% variation of the optical power (Table II in Supplement 1). This proves that the defects redistribute the power of the suppressed modes into the harmonic modes, partially impacting the overall device performance. However, since there is no clear correlation with the number of defects implemented, such performance variations between the different HFCs are mostly device dependent.

The Fourier transform infrared (FTIR) emission spectra (Bruker, Vertex 80) taken under the vacuum of the reference device



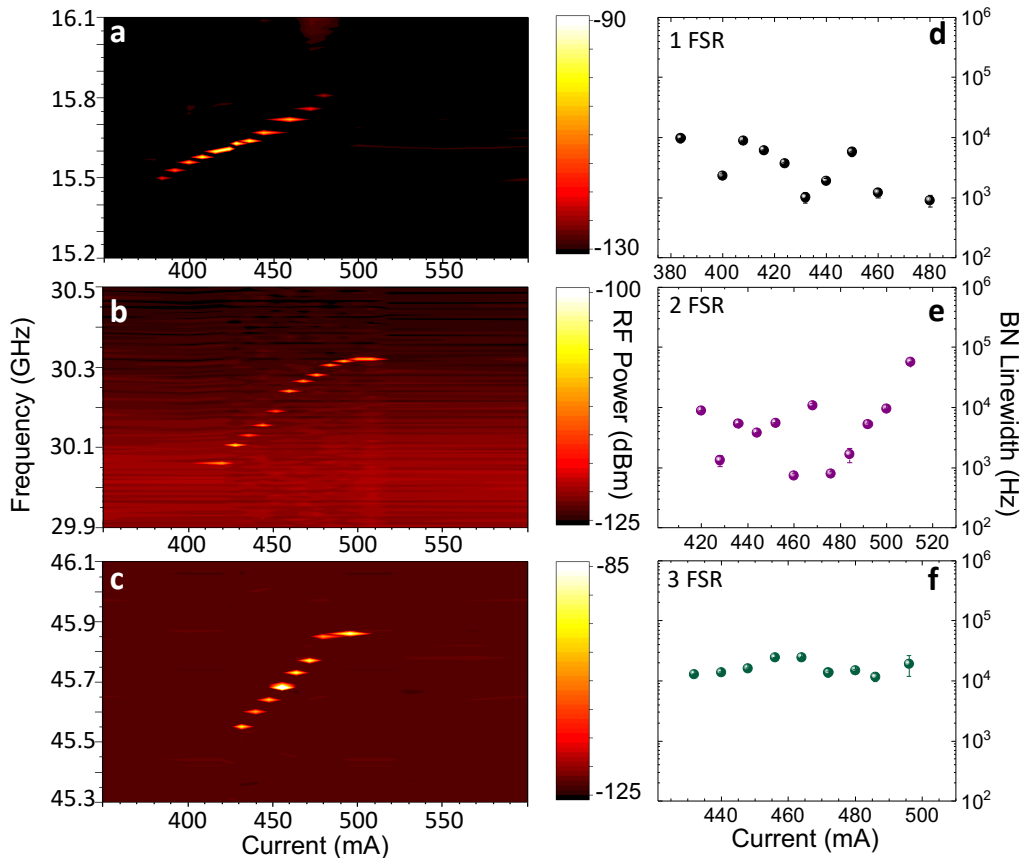


**Fig. 2.** Harmonic frequency comb spectra. (a) Scanning electron microscope (SEM) image of a double-metal QCL with the defects patterned on the top contact. (b) Voltage–Current density ( $V$ – $J$ ) and Light–Current density ( $L$ – $J$ ) characteristics of the HFC with  $n = 5$ , measured at 15 K. The optical power was measured using a broad-area THz absolute power meter (TK Instruments, aperture  $55 \times 40 \text{ mm}^2$ ). (c) The FTIR emission spectrum of a non-patterned reference QCL acquired under a vacuum in a continuous-wave (CW) at 20 K, with a  $0.075 \text{ cm}^{-1}$  spectral resolution. (d)–(f) The FTIR spectra of three devices with the same dimensions as the reference laser of panel (c) but with an increasing number of defects on the top contact [ $n = 1$  (d),  $n = 2$  (e), and  $n = 5$  (f)]. The driving currents, corresponding to the  $V$ – $J$  curve of panel (b), are reported on the graphs. The dashed lines mark the noise level. The spectra become perfectly harmonic according to the distance between the defects (2, 3, and 6 free spectral range (FSR)). All spectra are collected at 20 K. (g)–(i) Intermode beatnote of the reference laser (g) and of the device with  $n = 1$  (h) and  $n = 2$  (i), recorded with an RF spectrum analyzer [Rohde and Schwarz FSW43, RBW: 300 Hz; video bandwidth (VBW): 300 Hz; sweep time (SWT): 20 ms; RMS acquisition mode], under the same conditions of the spectra (c)–(e). (l) Threshold current density of the HFC regime as a function of the harmonic order (FSR). (m) Interferogram (IFG) of the harmonic comb with  $n = 5$  corresponding to the spectrum (f), with two magnified views, one close to zero and one close to the maximum delay of the FTIR, showing the preservation of its symmetry.

[Fig. 2(c)], without defects, shows a FC regime with a maximum spectral coverage of 0.79 THz with a set of 50 optical modes equally spaced by 1 FSR = 15.7 GHz. When defects are implemented on the top of the laser cavity, the FTIR spectra show [Figs. 2(d)–2(f)] a pure harmonic regime, with a repetition rate that precisely follows the rationale of our simulation design, e.g.,  $n$  defects correspond to a HFC with a mode spacing equal to  $(n + 1)$  FSR, as shown by the FTIR spectra and by the corresponding intermode beatnotes [Figs. 2(g)–2(i)].

In a free-running THz QCL, the FC regime is self-starting and persists only in specific operational regimes, usually just above the laser threshold [9,34,36], in which the group velocity dispersion [37–39] is low enough. The gain medium, however, entangles the dispersion dynamics at higher driving currents, resulting in the FC

operation usually occurring over less than 25% of the lasing bias range [33]. Without a strategy to handle such a bias-dependent dispersion compensation, which includes active and passive approaches [40–44] or strategies to modulate the intra-cavity field [10], the achievable spectral range of the FC is limited with respect to that set by the gain medium design. This is the case of the reference structure of the present work [Fig. 2(c)], in agreement with previous reports [10,34]. However, the devised HFCs show a different behavior: the current density threshold of the HFC regime  $J_{\text{HFC}}$  increases as a function of  $n$ , as shown in Fig. 2(l). This is in agreement with previous experimental reports in self-starting homogeneous THz HFCs [28] and consistent with the increased losses found at increasing  $n$  (see Supplement 1, Fig. S3c). Below  $J_{\text{HFC}}$ , the spectra appear mostly single mode.



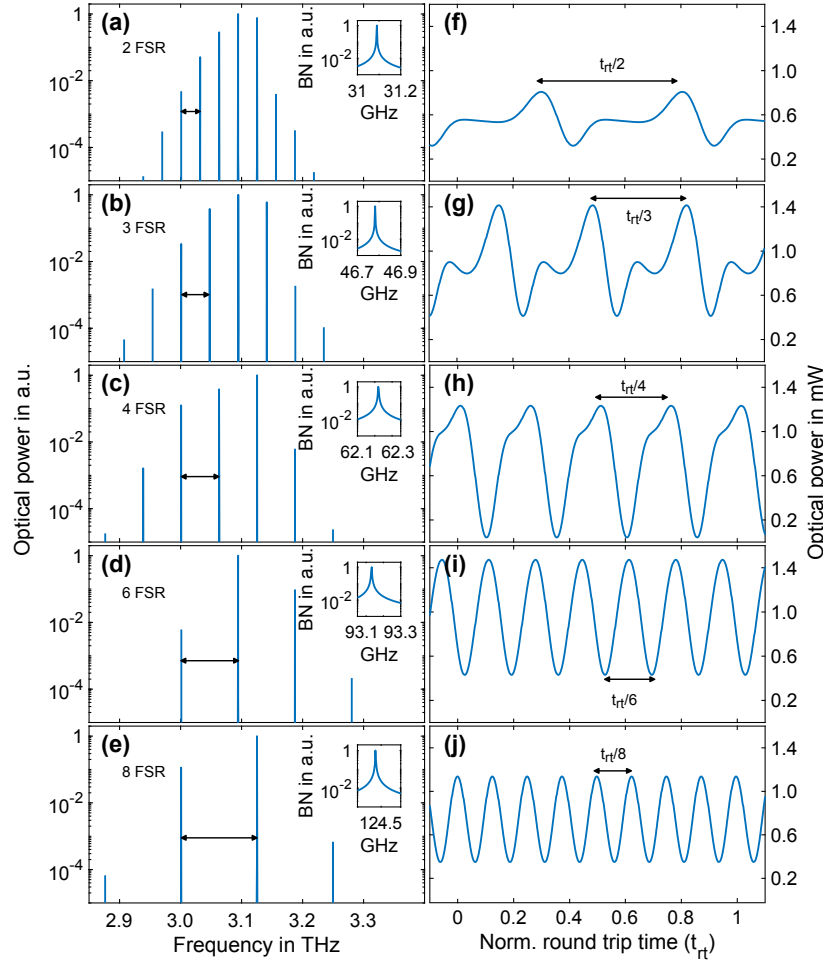
**Fig. 3.** Intermode beatnote (BN) maps and linewidth. (a)–(c) Intermode beatnote maps as a function of the driving current measured at 20 K, in continuous wave (CW), for (a) the QCL FC designed without defects, (b) with  $n = 1$ , and (c)  $n = 2$ . (d)–(f) BN linewidths as a function of the driving current measured at 20 K, in CW, for (d) the QCL FC designed without defects, (e) with  $n = 1$ , and (f)  $n = 2$ .

Interestingly, and in contrast to the reference laser, a stable HFC regime is found progressively more towards the driving current region in which the gain is clamped (530–750 mA) [34], demonstrating that the QCL operates as a stable self-starting FC without any of the aforementioned dispersion compensation strategies needing to be implemented. Here, the laser delivers its maximum optical power, and hence the spectral coverage becomes broader, turning from 0.8 THz [ $n = 1$ , Fig. 2(d)] to 0.85 THz [ $n = 2$ , Fig. 2(e)] and then to 1.1 THz [ $n = 5$ , Fig. 2(f)]. Remarkably, the device [Figs. 2(d)–2(f)] delivers a maximum optical power per mode of  $\sim 270 \mu\text{W}$ , one of the largest values reported so far in a THz QCL FC. This is reflected in the plots of the intermode beatnote (BN) as a function of driving current [Figs. 3(a)–3(c)] that we detected via a free space antenna using a radio frequency spectrum analyzer equipped with a 90 GHz mixer (Rohde and Schwarz FS-Z90) to extend its operational range up to 90 GHz. In the case of the reference laser [Fig. 2(c)], a single, sharp ( $\sim 40 \text{ dBm}$ ) and narrow linewidth (LW) of  $\sim 0.7 - 10 \text{ kHz}$  BN, at  $\sim 15.7 \text{ GHz}$ , persists for a current range of 100 mA (i.e., from 385 to 490 mA, 13% operational range), in agreement with previous reports [Fig. 3(a)] [34]. For the HFC with  $n = 1$ , the  $\sim 30.3 \text{ GHz}$  BN (2FSR), having a signal-to-noise ratio of 28 dBm, persists for  $\sim 100 \text{ mA}$  (i.e., from 420 to 520 mA) [Fig. 3(b)] and shows a narrow LW (1–10 kHz). The HFC with  $n = 2$  shows a stable and narrow (10–20 kHz) BN ( $\sim 35 \text{ dBm}$ ) at  $\sim 45.7 \text{ GHz}$  (3FSR) persisting over  $\sim 80 \text{ mA}$  (i.e., from 430 to 500 mA) [Fig. 3(c)]. It is worth mentioning that, in both cases, and in the investigated

current range, no other beating across the RF spectrum is observed except the one of the harmonic beatnote, indicating the purity of the harmonic state. This was proved by inspecting the RF spectra at the subharmonic frequencies with a lower resolution bandwidth (RBW) and a lower frequency span.

For the HFC with  $n = 5$ , we could not collect the corresponding BN due to the limitation of our spectrum analyzer. However, the coherence of the harmonic state can be verified to a certain degree by comparing the symmetry and periodicity of the measured interferogram envelope. Indeed, it has been shown [28] that preserving the symmetry in the interferogram in each period throughout the travel range of the FTIR is a necessary condition and an accessible tool to assess the presence of a pure comb state. Figure 2(m) shows the interferogram of the HFC with  $n = 5$ , corresponding to the spectrum 2f. The two magnified plots around the zero-path delay and close to the maximum travel range, respectively, show the required symmetry preservation.

The same device also exhibits a harmonic-like regime at 3 FSR that develops from 400 to 500 mA (see Supplement 1). This behavior is expected, considering the proposed design. In fact, the  $n = 5$  defects include the  $n = 2$  defects of the 3 FSR harmonic design. These two defects induce, in a separate current regime, the device to also develop this submultiple harmonic-like behavior before reaching the threshold of the sixth harmonic comb regime. In this regime, the spectrum appears nearly harmonic, and the corresponding interferogram does not preserve its symmetry throughout the travel range of the FTIR (see Supplement 1).



**Fig. 4.** Dynamical simulation results using the Maxwell-density matrix formalism. (a)–(e) Power spectra calculated from the last 5000 round trips for  $n = 1, 2, 3, 5,$  and  $7$  slits. The spacing between the individual comb lines closely follows the number of defects. Insets: RF beatnote (BN). The BN frequency increases proportionally with the order of the harmonic comb. The BN width is limited by the numerical frequency resolution ( $\sim 1.56$  MHz), indicating a phase-locked comb operation for each device. (f)–(j) Time traces of the outcoupled optical power for the corresponding spectra shown in the left column. Significant amplitude modulations are visible, with a periodicity corresponding to  $t_{rt}$  divided by the order of the harmonic comb.

**Table 1.** Simulation Parameters<sup>a</sup>

Parameter	Value
Loss coefficient $a_0$	$(12.75 + 0.25 \times n) \text{ cm}^{-1}$
Overlap factor	1
Center frequency	3.0 THz
Mirror reflectivity (power)	70.88%
Slit reflectivity (power)	0.04%
Refractive index	3.66
GVD coefficient $\beta_2$	$1.0 \times 10^{-22} \text{ s}^2 \text{ m}^{-1}$

<sup>a</sup>Waveguide parameters for the Maxwell-density matrix simulations.  $n$  denotes the number of defects.

#### 4. TIME DOMAIN SIMULATIONS

We next investigate the dynamical processes underlying harmonic comb generation by performing time-domain simulations, employing a Maxwell-density matrix approach [45–47]. The optical propagation equations for the waveguide field components are here coupled to a density matrix description of the active region dynamics (see Section 2). The waveguide parameters used can be found in Table 1.

In Figs. 4(a)–4(e), the simulated power spectra for  $n = 1, 2, 3, 5,$  and  $7$  equally spaced slits in the waveguide are shown along with the corresponding RF beatnote. As seen experimentally, the harmonic order of the comb directly scales with the number of slits, i.e., the comb line spacing and thus the RF beatnote frequency is related to the round-trip time  $t_{rt}$  via  $\Delta f = (n + 1)/t_{rt}$ . In all cases, narrow beatnotes with widths below the numerical resolution limit are obtained, indicating a phase-locked comb operation in agreement with the experimental results.

In Figs. 4(f)–4(j), the corresponding time traces of the outcoupled power are shown, featuring a significant amplitude modulation at the corresponding harmonic of the round-trip time. To obtain the harmonic combs of different orders shown in Fig. 4, we had to adapt the currents by slightly changing the doping.

For example, in Fig. 4(a), ( $n = 1$ ) was obtained for 466 mA, and in Fig. 4(e), ( $n = 7$ ) for 487 mA. This coincides with experimental observations that the current required for harmonic comb operation gradually increases with the number of slits. Also, the spectral coverage becomes broader, in agreement with above experimental results. For currents below the harmonic comb regime, our simulations yield single-mode lasing, and for currents above the harmonic comb regime, an unlocked multi-mode operation

with a broad beatnote is obtained, in agreement with the beatnote maps of Fig. 3. These results indicate that the defects introduce an additional resonance condition in the waveguide, favoring the respective modes and driving the field into a periodic waveform at the corresponding harmonic of the fundamental repetition rate.

For too high intensities, however, other mechanisms such as the Risken–Nummedal–Graham–Haken instability start to overcome the effect introduced by the slits, destroying the phase coherence between the modes [48].

## 5. DISCUSSION AND CONCLUSION

By implementing a defined number of defects on the top contact of a wire THz laser, we introduce a spatially periodic modulation of the optical gain, which promotes the emission of the HFC states. The purely harmonic regimes, developing at bias currents close to the peak of the laser emission, shows a spectral bandwidth significantly larger (1.1 THz) than that found on the free running fundamental harmonic (1FSR) THz QCL combs. The ability of the laser to distribute its optical power among a few powerful modes separated by many FSRs (as opposed to many, weaker adjacent cavity modes) opens up previously unforeseen possibilities for QCLs for arbitrary optical and microwave waveform generation [49] or for mode locking [50]. Indeed, provided that the repetition rate is on the order of the inverse of the gain recovery time or faster, the QCL can no longer be considered a fast saturable gain medium in the HFC regime, as was previously assumed [26]. This is also extremely appealing for broadband spectroscopy. The advantage of the HFC over the FC is that the higher power per mode and broader spacing characteristic of this state are expected to lead to a greater mode proliferation, and thus ultimately to a broader spectral coverage [50]. Furthermore, HFCs open intriguing perspectives for THz quantum key distribution of interest in quantum technologies or for the generation of terahertz signals of high spectral purity in wireless communication networks [13].

## APPENDIX A: METHODS

### 1. Active Region Design

The individual active regions consist of nine GaAs quantum wells, forming a cascade of alternating photon and LO-phonon-assisted transitions between two quasi-minibands [51]. Each active region module shows a diagonal laser transition. The layer sequences of the three active regions designs are: 10.6/**0.5**/17/1/13.5/**2.1**/12.4/**3.1**/10/**3.1**/9/**2.9**/7.5/**3.1**/17.8/**3.1**/15.2/**4.1**; 9.8/**0.5**/15.7/1/12.5/2/11.4/**2.9**/9.2/**2.9**/8.3/**2.9**/6.9/**2.9**/16.5/**2.9**/14.1/**3.9**; and 10.5/**0.5**/12.2/1/12.5/1.9/11/2.8/8.8/**2.8**/7.9/**2.8**/6.6/**2.8**/15.8/**2.8**/13.8/**3.7**, where the thickness is in nanometers, the bold figures represent the AlGaAs barriers, and the underlined figures denote the Si-doped ( $3.5 \times 10^{16} \text{ cm}^{-3}$ ) GaAs layer. The aluminum content in the barriers is 0.14, 0.16, and 0.18, respectively.

### 2. QCL Fabrication

Fabry–Perot laser bars are fabricated in a metal–metal waveguide configuration via Au–Au thermo-compression wafer bonding of the 17- $\mu\text{m}$ -thick active region (sample L1494) onto a highly doped GaAs substrate, followed by the removal, through a combination

of mechanical lapping and wet etching, of the host GaAs substrate of the molecular beam epitaxial (MBE) material. The  $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$  etch stop layer is then removed using HF etching. Vertical sidewalls are defined by inductively coupled plasma etching of the laser bars to provide uniform current injection. A Cr/Au (10  $\mu\text{m}$ /150  $\mu\text{m}$ ) top contact, patterned with  $2 \times 42 \mu\text{m}$  rectangular open slits, is then deposited along the center of the ridge surface, leaving a 3- $\mu\text{m}$ -wide region uncovered along the ridge edges. A thin layer of Ni (5-nm-thick) was then deposited over the defects and over the uncovered region (side absorbers) using a combination of optical lithography and thermal evaporation. This lossy Ni layer on the side absorbers is intended to inhibit lasing of the higher order lateral modes by increasing their threshold gain [52]. Finally, the backside of the substrate is lapped down to 150  $\mu\text{m}$  to allow a proper thermal management and enable operation in CW. Laser bars, 70  $\mu\text{m}$  wide and 2.5 mm long, are then cleaved and mounted on a copper bar, wire bonded, and then mounted onto the cold finger of a He continuous-flow cryostat.

### 3. Model

The harmonic comb operation is modeled using a coupled Maxwell-density matrix approach [45], which is combined with carrier transport simulations to avoid free fitting parameters [46]. This approach has been found highly suitable for quantitative modeling of mode-locked QCL operation [46,47]. Here, the quantum active region dynamics is captured by a Lindblad-type density matrix equation for a representative QCL period, supplemented by periodic boundary conditions [45,46]. Similarly, as for previous simulations of a short-pulse structure based on the same active region design, the eigenenergies, optical dipole moment of the laser transition, and scattering/dephasing rates are obtained from the carrier transport simulations of the 2.5 THz stack [46]. The resulting model system comprises nine energy levels. Employing the rotating wave/slowly varying the amplitude approximation, the optical field propagation is described by [45]

$$v_g^{-1} \partial_t E^\pm \pm \partial_z E^\pm = p^\pm - a E^\pm - i \frac{\beta_2}{2} \partial_t^2 E^\pm, \quad (\text{A1})$$

with the complex electric field amplitudes  $E^{(\pm)}(x, t)$  of the forward/backward traveling component. Here,  $z$  and  $t$  denote the position in the waveguide and time. Furthermore,  $v_g$  is the group velocity,  $p^\pm(z, t)$  represents the polarization term computed from the density matrix [45,46],  $a$  is the power loss coefficient, and  $\beta_2$  denotes the background group velocity dispersion (GVD) coefficient. The waveguide slits are modeled using reflection and transmission coefficients  $r$  and  $t$ , with

$$E_L^- = r E_L^+ + t E_R^-, \quad E_R^+ = r E_R^- + t E_L^+. \quad (\text{A2})$$

Here, the subscripts  $L$  and  $R$  refer to the field components directly to the left and right of the slit. Each simulation runs over more than 10,000 roundtrips to ensure convergence to a steady state.

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**Data availability.** The data and relevant computer codes supporting this study are available upon reasonable request from the corresponding author.



**Supplemental document.** See Supplement 1 for supporting content.

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