Applied Physics Letters

Widely tunable harmonic frequency comb in a quantum cascade laser

Cite as: Appl. Phys. Lett. **113**, 031104 (2018); https://doi.org/10.1063/1.5039611
Submitted: 09 May 2018 . Accepted: 08 July 2018 . Published Online: 17 July 2018

Marco Piccardo, Paul Chevalier, Sajant Anand, Yongrui Wang, Dmitry Kazakov, Enrique A. Mejia, Feng Xie, Kevin Lascola, Alexey Belyanin, and Federico Capasso

APPLIED PHYSICS LETTERS **113**, 031104 (2018)

Widely tunable harmonic frequency comb in a quantum cascade laser

Marco Piccardo,[1] Paul Chevalier,[1] Sajant Anand,[1,2] Yongrui Wang,[3] Dmitry Kazakov,[1,4] Enrique A. Mejia,[1,5] Feng Xie,[6] Kevin Lascola,[6] Alexey Belyanin,[3] and Federico Capasso[1,a)]
[1]*Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA*
[2]*Department of Physics, Wake Forest University, Winston-Salem, North Carolina 27109, USA*
[3]*Department of Physics and Astronomy, Texas A&M University, College Station, Texas 77843, USA*
[4]*Department of Information Technology and Electrical Engineering, ETH Zürich, 8092 Zürich, Switzerland*
[5]*University of Texas at Austin, Austin, Texas 78712, USA*
[6]*Thorlabs Quantum Electronics (TQE), Jessup, Maryland 20794, USA*

(Received 9 May 2018; accepted 8 July 2018; published online 17 July 2018)

Self-starting harmonic frequency combs in quantum cascade lasers exhibit skipping of several tens of longitudinal modes of the cavity, producing widely spaced frequency combs which may be used for a number of applications, such as the generation of high-spectral-purity microwave and terahertz tones. Under pure electrical injection, the spacing of such combs is fixed by fundamental laser parameters and can hardly be controlled. Here, we demonstrate that harmonic frequency combs in quantum cascade lasers can be induced by optical injection of an external seed provided by a tunable source. This scheme enables wide tunability of the harmonic comb spacing, allowing the skipping between 44 and 171 longitudinal modes in a single device. *Published by AIP Publishing.*
https://doi.org/10.1063/1.5039611

Since their first demonstration in recent years,[1] optical frequency combs in quantum cascade lasers[2–4] (QCLs) have been developed into a competitive technology for spectroscopy applications in the mid-infrared range.[5,6] Such QCL frequency combs are characterized by a spacing between longitudinal modes of one free spectral range (FSR), which is fixed by the device properties, namely, by the length of the cavity and effective group index of the waveguide.[7] A different type of QCL comb was recently discovered based on a new laser regime—the harmonic state[8,9]—originating from a single-mode instability transferring energy from the first lasing mode to optical sidebands separated by roughly the Rabi frequency of the primary mode. The relative magnitudes of the Rabi frequency and the FSR in QCLs are such that the comb spacing corresponds to several multiples of the FSR. The harmonic comb formation is enabled by a third-order optical nonlinearity[10] and is reminiscent of mode proliferation in optically pumped microresonators.[11,12]

The self-starting nature of harmonic combs in QCLs is interesting from the point of view of both applications and fundamental laser science, as it allows the generation of widely spaced combs by simple injection of electrical current in the device. The spacing of such combs is defined by fundamental parameters of the laser active region, namely, the gain recovery time and dephasing time, in addition to the geometry of the cavity.[8,10] However, fabricating a device with precise values of such parameters for a deterministic design of the comb spacing is a daunting task, which would require bandstructure and dispersion engineering.[4] Moreover, such an approach based on fundamental laser design would not allow an active control of the number of skipped cavity modes in an operating device. Here, we demonstrate by means of optical injection seeding that harmonic states with a wide range of spacings can be generated from a single device, between 0.34 and 1.32 THz (corresponding to 44 and 171 FSRs, respectively). The achieved tunability is particularly attractive in view of applications utilizing the coherent beats among the modes to produce spectrally pure tones at the frequency of the comb intermodal spacing.[13,14]

The device under study is a continuous wave, buried heterostructure, Fabry-Perot (FP) QCL emitting at a central wavelength of 4.5 μm[15] and having a 6 mm long cavity, a 5 μm width waveguide, a high-reflectivity (HR) coating on the back facet ($R \approx 1$), and an anti-reflective (AR) coating on the front facet ($R \approx 0.01$). The FSR of the device is 7.7 GHz. The QCL is driven with a low-noise current driver (Wavelength Electronics QCL LAB 2000), and its temperature is stabilized at 16 °C using a low-thermal-drift temperature controller (Wavelength Electronics TC5). The spectral evolution of this laser in the free-running mode was previously reported in Ref. 8 showing, as a function of the injected current, a transition from the single-mode regime to the sparsely populated harmonic state, and eventually at high current to a dense state, where adjacent cavity modes are populated. Upon pure electrical injection, the harmonic comb generated by this device exhibits a spacing of approximately 350 GHz.

As originally explained, the formation of the harmonic state can be triggered by a photon spontaneously emitted at a different frequency with respect to that of the first lasing mode.[8] This induces a beat note, i.e., an intensity modulation at the difference frequency of the two fields, and results in a parametric contribution to the gain of the spontaneous photon. At a pumping level known as the instability threshold, this parametric gain can allow two harmonic sidebands to overcome the losses and start lasing. Here, we demonstrate that the harmonic state can also be induced by the injection of an optical seed in the laser cavity, while the device is

a)capasso@seas.harvard.edu

operated below the instability threshold. A schematic of the set-up is shown in Fig. 1(a). The source of the optical seed is an external-cavity (EC) QCL (Daylight Solutions, model 41045-HHG) generating a single mode in continuous wave operation with a linewidth of 100 MHz. The seed is focused at an angle of 34° with respect to the normal to the AR facet of the FP-QCL, chosen to be larger than the half-divergence angle of the laser under injection. This configuration allows for the coupling of a fraction of the optical power of the seed into the cavity of the FP-QCL, while preventing the output of the latter to destabilize the operation of the EC-QCL, thus acting as an effective optical isolator. The coupling of the optical seed into the FP cavity depends on the relative detuning between the seed frequency and the nearest FP cavity mode, which can be varied by acting on different experimental parameters, as discussed below. The value of maximum coupling efficiency, i.e., close to zero detuning, is estimated to be 1.25% (see supplementary material for further details). The light emitted from the FP-QCL under injection is collimated by an off-axis parabolic mirror, and its emission spectrum is measured using a Fourier transform infrared spectrometer.

Prior to optical injection, the FP-QCL operates in the single mode regime [Fig. 1(b), bottom]. Injecting an optical seed detuned by 400 GHz with respect to the single mode frequency can induce the laser to evolve into either of two different states, a frequency-pulled single mode or a harmonic state [Fig. 1(b), middle and top]. In the first case, the frequency of the pulled single mode coincides with that of the seed, as typically observed in injection locking experiments.[16–18] In the other case, the central mode coincides with the single mode of the free running laser, while a sideband occurs at the frequency of the injected seed and another symmetric sideband is produced by four-wave mixing (FWM). The specific spectral evolution of the laser under optical injection depends on a number of experimental parameters. As shown in Figs. 1(c)–1(e), varying any of the quantities among FP-QCL current, FP-QCL temperature, and seed power can produce these two different types of states. [In few instances, only a weak sideband at the seed frequency is produced, apart from which the laser state remains essentially identical to the initial single mode. Such cases correspond to the missing points in Figs. 1(c)–1(e) and are due to a weak coupling of the seed into the QCL cavity, which is insufficient to generate an effect on the spectrum.] All these parameters play the same role, in the sense of producing a change in the group refractive index of the waveguide of the FP-QCL—either by changing its temperature directly (FP-QCL temperature) or by Joule heating (FP-QCL current) or by means of optical absorption (seed power). As a consequence, the FP modes of the cavity can be shifted with respect to the fixed frequency of the optical seed, varying the seed transmission into the cavity and affecting the spectral evolution of the laser. In a linear tuning regime, one may expect a periodic alternation among the different states as the parameters are progressively varied, due to the nearly uniform spacing of the cavity modes. Indeed, the two regimes of harmonic modes observed by temperature tuning are separated by a temperature difference [1.3 °C, Fig. 1(d)] corresponding approximately to the FSR of the cavity (7.8 GHz assuming a tuning coefficient[19] of $-8.9 \times 10^{-5}\,K^{-1}$). However, in the case of current tuning, the separation among the harmonic regimes [$0.03\,kA/cm^2$, Fig. 1(c)] corresponds only to a fraction of the FSR (1.8 GHz from the measured tuning coefficient of $60\,GHz\,cm^2\,kA^{-1}$) as the current may have a more complicated effect. Overall, the aperiodicity observed in the experiments [Figs. 1(c)–1(e)] suggests that the interaction of the seed with the cavity modes is nonlinear, thus

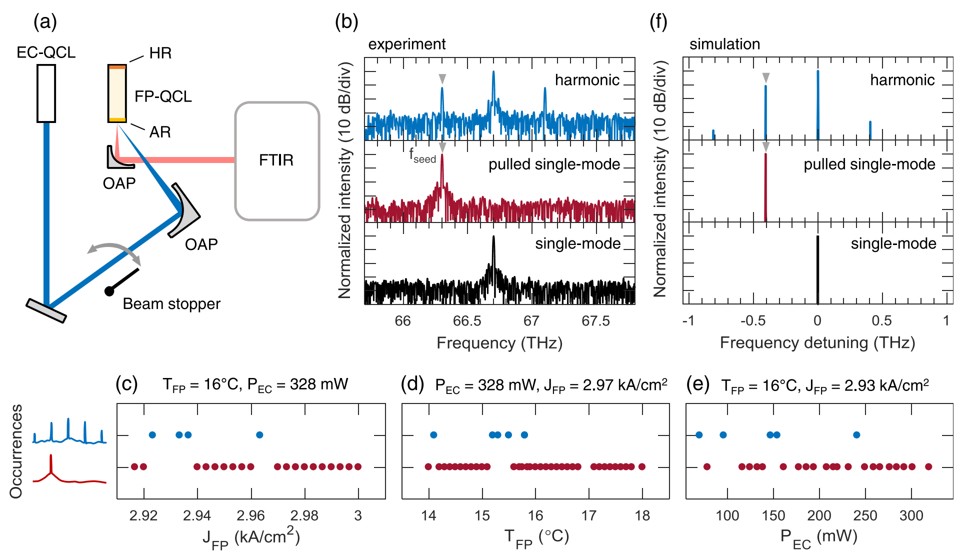

FIG. 1. Optically seeded quantum cascade laser. (a) Schematic of the experimental set-up used to induce harmonic comb operation in a Fabry-Perot QCL by injection of an optical seed generated from a tunable single-mode laser. (b) Prior to optical injection, the FP-QCL operates in the single mode regime (bottom). Upon injection of an optical seed (at a frequency marked by arrowheads), the laser can evolve into a frequency-pulled single mode (middle) or a harmonic state (top). The specific evolution depends on the following parameters: FP-QCL current density (J_{FP}) and temperature (T_{FP}) and incident EC-QCL seed power (P_{EC}). (c)–(e) Occurrences of the two different types of state shown in (b) as J_{FP}, T_{FP}, or P_{EC} are varied. (f) Space-time domain simulations of the QCL spectral evolution upon injection of an optical seed. Before optical injection, the laser operates in the single mode regime (bottom) and the amplitude of the reflected electric field at the AR facet of the laser is 1.2 kV/cm. When the amplitude of the injected seed overcomes a threshold of 0.7 kV/cm, a frequency-pulled single mode is produced (middle), while smaller amplitude values result in a harmonic state (top, seed amplitude: 0.2 kV/cm).

leading to a quasi-chaotic sequence of the resulting laser states. To investigate the mechanism determining the spectral evolution of the FP-QCL under optical injection in a more controlled framework, we resort to simulations of the experiment based on a space-time domain model of transport and recombination in the QCL active region[20] (see supplementary material for simulation parameters). It is found that when the amplitude of the coupled seed overcomes a threshold of 0.7 kV/cm (corresponding to a coupled power of 0.5 mW)—a value comparable to that of the FP-QCL mode reflected at the AR facet (1.2 kV/cm)—a frequency-pulled single mode is obtained [Fig. 1(f), middle]. The existence of a threshold is characteristic of an injection locking process.[7] On the other hand, for smaller values of seed amplitude, a harmonic state is observed, resulting from the amplification and mixing of the seed with the central mode of the FP-QCL [Fig. 1(f), top, seed amplitude: 0.2 kV/cm]. These results indicate that fine tuning the value of seed power coupled into the FP-QCL cavity is the key to control the generation of a harmonic state by optical injection. In particular, when the seed frequency lies within the locking range of a FP mode of the QCL cavity, a single mode is produced[21] due to the strong seed coupling and amplification, while outside of this range, the laser either evolves into a harmonic state or remains in the initial state due to the weak seed coupling. In addition to this, we note that in both experiments and simulations, the change of the laser state upon optical injection is reversible: turning off the seed sets the laser back into the initial single mode state, indicating that seed amplification is necessary to maintain the induced laser state.

In the following, we demonstrate that the harmonic comb spacing can be tuned by means of optical injection over a large frequency range, much beyond the intrinsic values. Moreover, we show that it is possible to induce a harmonic state not only in the case of a QCL operating in the single mode regime at low current but also starting from a dense multimode regime at high current, with the clear advantage of a larger optical output power. We start by driving the FP-QCL in the free-running mode at high current, where the optical output power is around 1 W and a dense state is produced with cavity modes separated by 1 FSR (Fig. 2, bottom). The injection of an optical seed destabilizes the multimode state. This can be intuitively understood as a result of a competition between the seed and the lasing modes for the extraction of gain from the active medium of the laser. By tuning the power of the incident optical seed, a variety of laser states can be produced inside the FP cavity (Fig. S2 in the supplementary material), including the harmonic state and a high-power single mode.[21] Its formation is explained as follows. The seeded mode has both a running-wave and a standing-wave component inside the HR/AR cavity of the FP-QCL. The seeded mode depletes the gain of all laser modes, either by consuming the population inversion at all spatial locations along the cavity with its running-wave component or by FWM processes, which extract energy from the lasing modes. Upon appropriate tuning of the seed power, all modes of the dense state become suppressed except the dominant one, which has access to more gain than other modes and is thus more stable. FWM between this dominant peak and the amplified seed results in the formation of a harmonic comb. To demonstrate the tunability of the comb spacing enabled by this approach, we present in Fig. 2(a) a series of spectra recorded from the injected FP-QCL exhibiting spacings between 0.34 and 1.32 THz, corresponding to 44 and 171 FSRs, respectively. The intensity of the generated sidebands is typically between 10 and 20 dB weaker than the central mode, a feature also observed in self-starting harmonic combs.[10] However, the optical power per mode is relatively high, being larger than 1 mW for all the visible sidebands in Fig. 2, thanks to the high-power (Watt-level) operation of the QCL. The limits to the tuning range of the harmonic comb spacing are set by the intrinsic properties and physics of the FP-QCL. The upper limit is given by the gain bandwidth of the laser, while the lower limit—very close to the spacing of self-starting harmonic combs observed in the device in free-running mode—is attributed to the nature of the parametric gain,[8] which

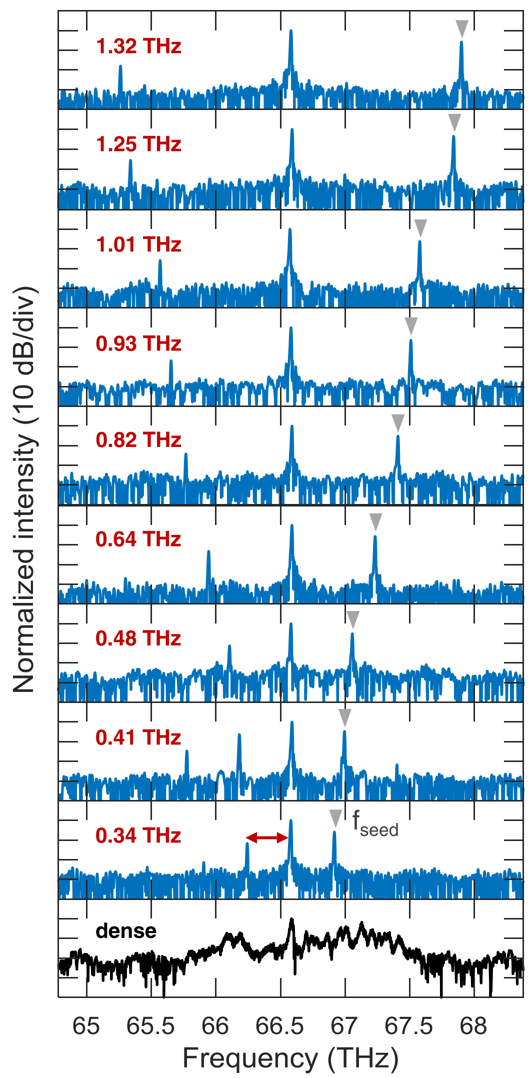

FIG. 2. Tuning the harmonic comb spacing in an optically seeded quantum cascade laser. Optical spectra emitted by the FP-QCL before (black curve) and after (blue curves) optical injection of seeds at different frequencies. The FP-QCL is operated at a current density of 3.59 kA/cm^2. By varying the seed frequency (marked by arrowheads), the harmonic comb spacing can be varied between 0.34 and 1.32 THz, corresponding to 44 and 171 FSRs, respectively.

tends to suppress low frequency modulations (see supplementary material for further details).

In terms of comparison with other systems, we note that FWM has also been demonstrated in quantum cascade amplifiers,[22] where all the optical sources are external and consist of two injected beams acting as the pump and signal for the FWM process. The advantage of the experimental scheme presented in this work is that we utilize an electrically injected FP-QCL, providing an internal source of optical power for FWM, while the injected optical beam acts as a relatively weak seed, allowing us to drive the evolution of the multimode field. Another appealing system where harmonic frequency comb generation with a THz spacing has been demonstrated is represented by optically pumped high-Q micro-ring resonators. By dual-pump schemes in such micro-cavities, wide harmonic combs with tens of skipped cavity modes were generated, exhibiting spacings between 0.29 and 2.25 THz (corresponding to 6 and 46 FSRs, respectively).[23] In the case of microresonators, particular care has to be taken to stabilize the frequency comb generation in order to eliminate the locking instability arising between the external pump and the cavity resonances—a significant experimental complication not occurring in optically injected QCLs.

The optical injection scheme demonstrated here allows us to exploit the full potential offered by the harmonic state of QCLs, enabling a controlled variation of the number of skipped longitudinal modes in the laser. This approach, namely, the use of an optical seed to drive parametric oscillations of the QCL, evocates the operation of microresonators, further highlighting the deep connection between these two comb platforms.[8] In the future, by exploiting the beating of the comb modes, harmonic frequency combs in QCLs may be used as tunable, coherent microwave and THz sources characterized by high-spectral-purity and room temperature operation.

See supplementary material for the supporting content.

This work was supported by the DARPA SCOUT program through Grant No. W31P4Q-16-1-0002. We acknowledge support from the National Science Foundation under Award No. ECCS-1614631. Any opinions, findings, conclusions, or recommendations expressed in this material are those of the authors and do not necessarily reflect the views of the Assistant Secretary of Defense for Research and Engineering or of the National Science Foundation.

[1] A. Hugi, G. Villares, S. Blaser, H. C. Liu, and J. Faist, Nature **492**, 229 (2012).
[2] J. Faist, G. Villares, G. Scalari, M. Rosch, C. Bonzon, A. Hugi, and M. Beck, Nanophotonics **5**, 272 (2016).
[3] D. Burghoff, T.-Y. Kao, N. Han, C. W. I. Chan, X. Cai, Y. Yang, D. J. Hayton, J.-R. Gao, J. L. Reno, and Q. Hu, Nat. Photonics **8**, 462 (2014).
[4] G. Villares, S. Riedi, J. Wolf, D. Kazakov, M. J. Süess, P. Jouy, M. Beck, and J. Faist, Optica **3**, 252 (2016).
[5] P. Jouy, J. M. Wolf, Y. Bidaux, P. Allmendinger, M. Mangold, M. Beck, and J. Faist, Appl. Phys. Lett. **111**, 141102 (2017).
[6] G. Villares, A. Hugi, S. Blaser, and J. Faist, Nat. Commun. **5**, 5192 (2014).
[7] A. E. Siegman, *Lasers* (University Science Books, 1986).
[8] T. S. Mansuripur, C. Vernet, P. Chevalier, G. Aoust, B. Schwarz, F. Xie, C. Caneau, K. Lascola, C-E. Zah, D. P. Caffey, T. Day, L. J. Missaggia, M. K. Connors, C. A. Wang, A. Belyanin, and F. Capasso, Phys. Rev. A **94**, 63807 (2016).
[9] M. Piccardo, P. Chevalier, T. S. Mansuripur, D. Kazakov, Y. Wang, N. A. Rubin, L. Meadowcroft, A. Belyanin, and F. Capasso, Opt. Express **26**, 9464 (2018).
[10] D. Kazakov, M. Piccardo, P. Chevalier, T. S. Mansuripur, Y. Wang, F. Xie, C. en Zah, K. Lascola, A. Belyanin, and F. Capasso, Nat. Photonics **11**, 789 (2017).
[11] T. J. Kippenberg, R. Holzwarth, and S. A. Diddams, Science **332**, 555 (2011).
[12] T. Herr, K. Hartinger, J. Riemensberger, C. Y. Wang, E. Gavartin, R. Holzwarth, M. L. Gorodetsky, and T. J. Kippenberg, Nat. Photonics **6**, 480 (2012).
[13] M. Tani, O. Morikawa, S. Matsuura, and M. Hangyo, Semicond. Sci. Technol. **20**, S151 (2005).
[14] T. Nagatsuma, G. Ducournau, and C. C. Renaud, Nat. Photonics **10**, 371 (2016).
[15] F. Xie, C. Caneau, H. P. LeBlanc, N. J. Visovsky, S. C. Chaparala, O. D. Deichmann, L. C. Hughes, C. e Zah, D. P. Caffey, and T. Day, IEEE J. Sel. Top. Quantum Electron. **17**, 1445 (2011).
[16] M. S. Taubman, T. L. Myers, B. D. Cannon, and R. M. Williams, Spectrochim. Acta, Part A **60**, 3457 (2004).
[17] C. Juretzka, H. Simos, A. Bogris, D. Syvridis, W. Elsäßer, and M. Carras, IEEE J. Quantum Electron. **51**, 1 (2015).
[18] A. Bogris, A. Herdt, D. Syvridis, and W. Elsäßer, IEEE J. Sel. Top. Quantum Electron. **23**, 1500107 (2017).
[19] A. Wittmann, M. Giovannini, J. Faist, L. Hvozdara, S. Blaser, D. Hofstetter, and E. Gini, Appl. Phys. Lett. **89**, 141116 (2006).
[20] Y. Wang and A. Belyanin, Opt. Express **23**, 4173 (2015).
[21] P. Chevalier, M. Piccardo, S. Anand, E. A. Mejia, Y. Wang, T. S. Mansuripur, F. Xie, K. Lascola, A. Belyanin, and F. Capasso, Appl. Phys. Lett. **112**, 061109 (2018).
[22] P. Friedli, H. Sigg, B. Hinkov, A. Hugi, S. Riedi, M. Beck, and J. Faist, Appl. Phys. Lett. **102**, 222104 (2013).
[23] W. Wang, S. T. Chu, B. E. Little, A. Pasquazi, Y. Wang, L. Wang, W. Zhang, L. Wang, X. Hu, G. Wang, H. Hu, Y. Su, F. Li, Y. Liu, and W. Zhao, Sci. Rep. **6**, 28501 (2016).