TOPICAL REVIEW

Interband cascade laser frequency combs

Lukasz A Sterczewski, Mahmood Bagheri, Clifford Frez, Chadwick L Canedy, Igor Vurgaftman, Mijin Kim, Chul Soo Kim, Charles D Merritt, William W Bewley and Jerry R Meyer

1 Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109, United States of America
2 Naval Research Laboratory, Washington, DC 20375, United States of America
3 KeyW Corporation, Hanover, MD 21076, United States of America

* Author to whom any correspondence should be addressed.
E-mail: Mahmood.Bagheri@jpl.nasa.gov

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Abstract

Interband cascade lasers (ICLs) have emerged as efficient, room-temperature semiconductor light sources with relatively high wallplug efficiency and low power consumption in the 3–6 µm wavelength region. The recent discovery that ICLs can generate self-starting optical frequency combs has triggered a new avenue of research that ultimately promises to provide broadband, gap-free chemical sensing sources for the simultaneous detection of multiple species or rapid scanning of broad absorption features. Here we review ICL frequency combs for the first time, focusing on device topologies, spectral characteristics, and frequency stabilization. Future perspectives such as the development of on-chip dual-comb spectrometers based on the ICL platform, real-time probes of molecular dynamics, and combs that use harmonic comb states to generate millimeter waves are also discussed.

1. Introduction

Miniaturized optical frequency comb (OFC) sources [1] are revolutionizing the landscapes of precision measurements and optical sensing [2]. The OFC’s unique pattern of narrow, equidistant, phase-locked optical lines is advantageous in many applications because the frequency of each ‘tooth’ in the comb can be quantified by two microwave frequencies: a common offset \( f_0 \), and the repetition rate \( f_{\text{rep}} \), both of which are lockable to metrological standards [3, 4]. Compared to single-frequency lasers, OFCs offer enormous optical bandwidth that can substantially exceed the intrinsic tunability of a typical laser diode, combined with straightforward calibration of the optical frequency. While the first OFCs relied on optically-pumped near-infrared (NIR) solid state or fiber lasers with megahertz repetition rates [3, 5], chip-scale semiconductor OFCs now offer electrically-pumped gigahertz-rate sources that span the near to far-infrared spectral regions [6]. Although semiconductor combs still struggle to replicate the noise performance and spectral coverage of the more mature platforms, their small footprint and ease of operation make them preferable for applications requiring high (milliwatt) power per line, low electrical drive power (<1 W), and rapid current tunability.

Whereas mature optically-pumped frequency combs with full on-chip integration capability exist in the NIR region [7–10], optically-pumped midwave infrared (MWIR) platforms such as those exploiting microresonators [11] are still pumped by bulky external optical parametric oscillators. This makes the potential for electrically-pumped OFCs with self-starting intracavity comb generation particularly attractive. Despite many technological challenges, MWIR spectroscopy is especially powerful because many environmentally-important molecular species have strong ro-vibrational MWIR transitions, which are difficult to measure using conventional tunable laser spectrometers. In particular, the wide spectral coverage and modal spacing of OFCs can probe the broadband spectra of heavier and more complex hydrocarbons such as volatile organic compounds.

Up to now, the leading electrically-pumped MWIR OFC source has employed the unipolar quantum cascade laser (QCL) gain medium [12], in which electrons cascade through multiple (typically 20–40) active stages of epitaxially-grown quantum wells (QWs) [13]. In contrast to a conventional diode laser, whose
wavelength is constrained by the bandgap of the quantum heterostructure, QCLs rely on engineered intersubband transitions that allow the same material system (usually InP-based) to offer broad spectral flexibility. QCL combs [12], which are now commercially available, display unmatched performance in the longwave infrared (LWIR) spectral region (7–9 µm) and have achieved the first far-infrared (THz) OFC operation [14]. Nonetheless, QCL comb optimization remains challenging, due to a large thermal load and difficulties managing the spectral dispersion [15].

Alternatively, the MWIR niche (e.g. 3 µm < λ < 6 µm) can be spanned by the interband cascade laser (ICL) [16–18]. An ICL merges the concepts of photon generation by interband electron–hole recombination, as in a conventional diode laser, with the QCL’s staircase of active stages that allow a single injected electron to emit multiple photons. Since an ICL provides more gain per stage than a QCL, fewer stages are needed (typically 3–10) to overcome optical losses in the cavity. Furthermore, the ICL’s upper-state lifetime is much longer than in a QCL, so lasing can be achieved with much lower threshold current density. Consequently, the typical ICL power budget is more than an order of magnitude lower than for a QCL [19, 20]. This advantage contributed to the remarkable success of the single-mode ICL deployed aboard the Curiosity Rover that detected methane on Mars [21, 22].

It is therefore not surprising that the ICL platform is promising for a new generation of MWIR OFCs with properties attractive for terrestrial and space applications. The first ICL combs were quite recently demonstrated experimentally [23–25] and discussed theoretically [26]. The best-performing devices employ a two-section cavity geometry, in which the short section is independably biasable, as shown schematically in figure 1(a). Figures 1(b)–(e) show the ridge waveguide cross-section along with an SEM picture and micrographs of an ICL comb mounted on a microwave-compatible BeO submount. The characterization data in figure 2 indicate that this remarkably small (several mm × several µm) device emits ∼10 mW of continuous wave (CW) optical power when operated at room temperature, while consuming <1 W of electrical power (figure 2(a)). Sub-THz-wide MWIR OFCs (figure 2(b)) will be ideal for demanding dual-comb spectroscopy (DCS) applications that require extremely narrow optical linewidths and high modal coherence.

This paper reviews the emerging ICL OFC technology for the first time. Section 2 discusses the comb characteristics, spectral evolution, and device topologies, and summarizes comb demonstrations in different spectral regions. Section 3 considers frequency stabilization via microwave injection locking and its effect on the optical spectrum, while section 4 treats tunability with injection current for gapless measurements. Section 5 discusses the mode-skipping phenomenon that induces harmonic comb emission in many devices. Finally, section 6 highlights applications in spectroscopy, particularly using the dual-comb configuration, followed by a short future perspective for ICL comb devices and their comparison with other on-chip frequency comb platforms.
2. Basics of ICL frequency comb generation

2.1. Frequency modulation (FM) comb operation

In the frequency domain, an OFC emits sharp, equidistant lines with well-defined intermodal phases. One way to produce such emission from a diode laser is by splitting the contact to create a saturable absorber (SA), which passively mode-locks a train of short optical pulses [27]. That is not the only solution, however, since it was observed even in the early days of semiconductor lasers that bulk, single-section Fabry–Pérot devices can produce narrow microwave beat notes that indicate some degree of mode synchronization within the cavity [28]. The phenomenon was often referred to as ‘passive FM locking’ [28], or ‘single-section passive mode locking’ [29]. Many semiconductor laser classes, including quantum dash (QDh) [30], quantum dot (QD) [31], and QW [28, 32, 33] lasers, plus more recently QCLs [12] and ICLs [24, 34], have demonstrated such behavior. It was concluded that intrinsic gain nonlinearities can overcome the cavity dispersion to synchronize the modal phases.

This state was often characterized by high group velocity dispersion (GVD) and quasi-CW optical power. The corresponding nonlinear intensity autocorrelation traces lacked the 8:1 peak-to-background ratio that signifies full passive mode-locking [35]. In some cases, however, an optical fiber could compensate the chirp and provide pulsed emission, as confirmed by the proper interferometric autocorrelation trace expected for passive mode-locking [32]. More recently, novel linear interferometric techniques such as shifted wave interference Fourier transform spectroscopy (SWIFTS) [36] have improved the understanding of this phenomenon in MWIR OFCs.

Following the discovery [37] and proof [12] of OFC generation by QCLs, extensive research concluded that comb formation in a medium with fast gain dynamics must be dominated by self-starting FM [38]. Because the QCL’s upper-state lifetime is very short (on the order of picoseconds) compared to the cavity roundtrip time, it cannot efficiently store enough energy for pulse generation and passive mode locking [39]. On the other hand, the longer lifetime associated with interband (electron–hole) optical transitions should in principle make the ICL an excellent candidate for passive mode locking via the on-chip integration of an SA.

2.2. ICL comb demonstrations

In 2017, Sterczewski et al observed the first intrinsic comb-like emission from single-section Fabry–Pérot ICLs, and demonstrated multiheterodyne spectroscopy in a dual-comb configuration [23, 24]. Emission spectra from the dual 2 mm long devices were centered at 3050 cm$^{-1}$ ($\lambda = 3.28$ µm), with 19.2 GHz repetition rates mismatched by 96 MHz. The multi-heterodyne beating of the cross-correlated electric fields of the two combs [40] lacked short bursts, suggesting highly-chirped emission similar to that from FM QCLs and relying on intrinsic gain nonlinearities. The bandwidth in this first comb demonstration was only 10 cm$^{-1}$ (300 GHz), due to mode-grouping and the lack of any dispersion management.

In 2018, JPL and NRL demonstrated ICL combs that combined independently-biasable gain and SA sections on the same laser ridge [25]. As shown in figure 2, these more optimized two-section OFCs displayed broader bandwidths (up to 30 cm$^{-1}$, or $\sim$1 THz) with $\sim$9.7 GHz repetition rates. The inset to figure 2(b) shows that the radio frequency (RF) beatnote was quite narrow (1 kHz). The short carrier lifetime
Table 1. Summary of ICL frequency comb demonstrations across spectral regions.

| Frequency range \((\text{cm}^{-1})^a\) | Wavelength range \((\mu\text{m})\) | Repetition rate (GHz) | Single/two-section design | DCS/ multiheterodyne | Reference |
|---------------------------------|---------------------------|-----------------|------------------|-------------------|----------|
| 3103–3120                      | 3.205–3.222               | 19.2            | Single-section FP | ✓                 | Sterczewski et al [24] |
| 2740–2780                      | 3.597–3.650               | 9.7             | Two-section with SA | ✓                | Bagheri et al [25], Sterczewski et al [41, 42, 50] |
| 2570–2610                      | 3.831–3.891               | 10.2            | Two-section FP    | ×                 | Schwartz et al [34] |
| 2575–2620                      | 3.817–3.883               | 10.2            | Two-section + RF | ×                 | Hillbrand et al [45] |
| 3082–3115                      | 3.210–3.245               | 13.2            | Two-section       | ✓\(^b\)          | Feng et al [46, 49] |
| 3570–3680                      | 2.72–2.80                 | 12.8            | Two-section       | ×                 | Feng et al [49] |

\(^a\) \(1\:\text{cm}^{-1} \approx 30\:\text{GHz}\).

\(^b\) Stabilized with delayed optical feedback.

in the SA (reduced by ion bombardment) was expected to promote pulsed emission with amplitude-modulated (AM) characteristics. However, the interferometric autocorrelation trace resembled that of an FM comb, rather than displaying the 8:1 peak-to-background ratio expected for AM devices [35]. Subsequent dual-comb experiments confirmed the lack of optical pulses [41, 42]. Whereas AM combs may be viewed as amplitude solitons similar to those encountered in fiber lasers, FM states correspond to phase solitons [43] with frequency modulated linearly in time.

The FM nature of two-section ICL combs (with the short section positively biased) was confirmed experimentally by Schwartz et al in 2019 [34] (TU Wien). Those authors used the short section for tunable dispersion compensation rather than as an SA. The comb characteristics resembled those encountered in QCLs, with intermodal phases spanning a full \(2\pi\) range [44]. Strikingly, when strong microwave modulation was applied to a similar device that was optimized for low RF loss, the comb generation switched from FM to AM operation [45]. Applying >1 W of microwave power to the modulator section synchronized the intermodal phases via active mode locking, which led to picosecond MWIR pulse emission. The reluctance of ICL combs to display passive mode-locking, even under strong negative bias of the SA section, remains unclear. The Auger-limited carrier lifetime is 100–200 ps at the typical operating point of the gain section, with the differential lifetime a factor of 3 shorter. The fact that it is comparable to the cavity round-trip time [46] is expected to reduce the pulse peak power, but is unlikely to prevent the pulse formation by passive mode locking, as in the case of the QCL. It is also possible that the vertical carrier extraction from the SA section is too slow even at significant negative biases if the SA section is not ion bombarded to reduce the lifetime sufficiently [47]. Intracavity dispersion is also quite high in typical ICL designs, with typical values in the 1500–2500 fs\(^2\) mm\(^{-1}\) range [45, 48]. While further research is needed to pinpoint the origin of problems with pulse formation, whenever such problems exist, FM locking becomes the default mechanism underlying牙齿 synchronization even in two-section devices.

Although we focus primarily on type-II ICL combs in this review, it should be noted that an alternative interband cascade OFC platform has emerged recently, pioneered by researchers at State University of New York (SUNY) at Stony Brook. Two-section interband cascade devices employing active transitions in type-I QWs have demonstrated true passive mode locking under strong negative bias to the absorber section [46], which was confirmed by nonlinear interferometry. Type-I ICL OFCs have displayed attractive properties in the spectroscopically-relevant regions of 3.2 \(\mu\text{m}\) and 2.7 \(\mu\text{m}\) [49], and they show compatibility with dual-comb configurations when stabilized using delayed optical feedback. Table 1 summarizes the ICL combs that have been reported to date.

### 2.3. Optimizing an FM ICL comb

Because the intrinsic emission characteristics of all the type-II ICL combs reported to date have indicated FM operation, we will briefly review the key prerequisites for generating a stable FM comb. The first is multi-mode operation, which occurs when spatial hole burning (SHB) induces gain non-uniformities by creating carrier gratings inside the cavity [51]. Just above threshold, SHB more easily fulfills this requirement in an ICL, with longer wavelength, than in a near-IR laser [52]. Next, four-wave-mixing (FWM) has been identified as the primary mechanism locking the modal phases [51], which are otherwise non-equidistant as governed by the cavity’s GVD. The efficiency of this third-order nonlinear optical process increases at higher optical intensities, although it may still fail to compensate an extreme GVD if the spectrum becomes too broad at high injection currents. Furthermore, FWM alone may be insufficient to induce multimode
Figure 3 shows the optical spectra for various operating regimes of a typical FM ICL comb with a 4 mm long cavity. The laser was thermally stabilized at 25°C, and biased using a low-noise current driver. Light from the facet was collimated by a short-focal-length black diamond lens, and guided into a Fourier transform infrared (FTIR) spectrometer. Simultaneously, the microwave intermode beat note was extracted electrically from the cavity's short section. This signal may be viewed as a result of modal 'self-mixing' due to optical and electrical nonlinearities of the ICL.

Intuitively, we expect a sharp and clean microwave tone when all the generated lines are equidistant and tightly locked, whereas a pronounced noise pedestal suggests imperfect phase coherence for some of the lines. It should be noted, however, that while a clean microwave tone provides one indication of comb operation, it should not be considered proof. This is because as few as two lines can produce a spectrally-narrow signal, even when the rest are completely incoherent. More definitive confirmation requires either a spectrally-resolved measurement of the comb’s coherence using the SWIFTS technique, or the observation of optical beating with a second OFC.

As illustrated in figure 3, ICL combs typically enter a multimode regime shortly above threshold, which produces spectrally-rich yet narrow-bandwidth combs. However, as the comb broadens a pronounced noise pedestal often appears, due to more difficult compensation of the intrinsic GVD. The most stable comb regime is observed next, at intermediate (2–3.5 times threshold) current injection, albeit with high modal sparsity. At still higher currents the comb enters a second dense regime, although this time with a broad (multi-MHz-wide) beat note that widens even more when a second lobe appears at longer wavelength. The latter phenomenon is attributable to modal leakage into the high-index GaSb substrate [48], which quasi-periodically modulates the laser gain and GVD. Similar characterization of single-section devices operating around 3.2 µm revealed that this state with high phase noise and multiple spectral lobes is accompanied by pronounced low-frequency (near DC) intensity noise [23]. This may result from rectification of the microwave intermode beat note, due to electrical nonlinearities and thus conversion to baseband. From a practical standpoint, it may play an important role in the comb diagnostics. Whereas looking at the GHz-frequency microwave beat note is a standard and well-established technique to identify comb regimes, a wealth of knowledge about ICL comb stability may be inferred from radio frequencies up to ∼10 MHz, which are conveniently accessible using much slower (and cheaper) electronics.

This example has illustrated general characteristics of the platform, even though the current and temperature ranges over which the different regimes appear may vary from device to device. Interestingly, it is not only two-section ICL combs that enter the sparse yet stable comb regime, since [24] reported a similar phenomenon in symmetric uncoated FP devices. Furthermore, mode-grouping effects that limit the comb bandwidth have been observed across different devices, and are currently being analyzed in detail [48].

3. Injection locking for frequency stabilization

Although the free-running stability of an ICL comb is excellent, with repetition rate fluctuations $\Delta f_{\text{rep}}/f_{\text{rep}}$ on the order of $10^{-7}$ and optical linewidths <400 kHz over millisecond scales, many scenarios require
frequency stabilization. For instance, sensing systems that exploit enhancement in an optical cavity do not tolerate laser frequency drift. We may also want a frequency standard to define the repetition rate. Note that this level of free-running stability is only slightly less than for mid-infrared QCL combs, where the $\Delta f_{\text{rep}}/f_{\text{rep}}$ is on the order of $10^{-8}$ and optical linewidths are <100 kHz [54]. This difference, which may be attributed to the high positive GVD of current ICLs, perturbs phase locking and will be addressed in future designs.

The mechanism that enables repetition rate stabilization via injection locking depends on the type of comb and cavity geometry. In two-section devices, the short section can be seen as a fast intensity modulator, which promotes (stabilizes) the mode spacing defined by the external RF tone via gain/loss mechanisms. At extreme levels of injected power (e.g. >1 W), it can also actively lock the optical phases to produce pulsed emission (AM comb) [45]. However, lower RF signal levels (~mW) simply stabilize the repetition rate without affecting the comb’s modal and temporal profiles, as will be discussed below. In the case of a single-section cavity, an injected microwave signal can produce a microwave grating (standing wave pattern) that spatially and temporally modulates the injection current. The same effect has been observed to play a role in locking two-section cavities as well [54].

Figure 4 shows the experimental setup and free-running characteristics of a two-section ICL comb whose low-capacitance short section is well suited for this purpose [42]. Furthermore, for optical detection of the intermode beat note under microwave locking conditions, the ICL medium’s bi-functionality [34, 42, 55] was exploited to fabricate an interband cascade detector (ICD) from the same material as the comb laser.

The RF signal for microwave injection [56] was applied to the laser short section in two scenarios. In the first, the RF power was fixed while the injection frequency was swept symmetrically around the natural roundtrip frequency (figure 5(a)), whereas in the second the power was varied with the injected frequency held fixed at that of the free-running laser (figure 5(b)). At 6 dBm of RF power, locking was maintained over a 6 MHz range. Figure 5(b) shows that even at small detuning from the natural frequency, a minimum RF power is required to suppress the sidebands (<−5 dBm) and a slight noise pedestal (<5 dBm). Figure 5(c) confirms that the moderate injection levels which yield a clean microwave tone over the entire locking range have little effect on the optical spectrum. Figure 5(d) shows that the dependence of locking range on injected power follows the square root model [57]. TU Wien also used RF injection to lock the ICL comb frequency [34], and to obtain pulse lengths as short as 3.2 ps [45].

Unfortunately, it is challenging to independently lock the offset frequency $f_0$ of a semiconductor OFC, in contrast to pulsed fiber lasers. The gold standard of frequency comb stabilization, the self-referencing $f−2f$ technique [3], cannot be implemented here because ICL combs cannot attain the short pulses required to broaden bandwidths to an octave. From a practical standpoint, the offset frequency can be retrieved and locked by heterodyning against a second laser with known frequency. Comb frequencies can also be referenced to a temperature-stabilized optical cavity, or a low-pressure gas absorption line. Such techniques can lock the absolute frequency, while relative offset locking between two combs (without anchoring to a
Figure 5. Microwave injection locking experiment. (a) Injection locking map—measured RF spectrum as a function of detuning from the free-running repetition rate at constant injection power. (b) Evolution of the microwave beat note spectrum for constant detuning from the natural repetition rate as a function of injected power. (c) Optical spectrum with the +5 dBm RF injection On (red) and Off (blue), proving negligible influence. (d) Experimental locking range as a function of injected power (points), along with a square root fit (curve) \[57\].

Figure 6. Full-FSR tuning, enabled by injecting 0 dBm of microwave power into the ICL cavity. The laser current was incrementally varied over 50 steps between 207.5 and 218.5 mA. Although the modal intensities changed somewhat, the trajectory was largely reproducible. The \(\sim 100\) comb lines generated in this manner can replace an array of tunable single-mode lasers (previously-unpublished data).

4. Gap-free tuning

A remarkable feature of semiconductor laser OFCs is their rapid tunability via injection current, which can fill in the mode spacing to obtain gapless measurements \[62\]. Typical current tuning coefficients are on the order of \(\sim 1\) GHz mA\(^{-1}\), although given the different comb regimes and changes in spectral envelope, it is not always possible to scan the full free spectral-range (full-FSR). Although slower, temperature tuning with typical temperature coefficients on the order of 0.3 cm\(^{-1}\) K\(^{-1}\) (9 GHz K\(^{-1}\)) generally imposes less qualitative effect on the comb states. The microwave injection locking described in the previous section can also provide gapless tuning, by locking the laser to a particular comb regime for longer than when the laser is free-running. Figure 6 shows the spectral evolution of an ICL comb subject to 0 dBm (1 mW) of microwave signal, when the injection current was incrementally varied over 11 mA in 50 steps. The result was full-FSR tuning with only minor changes in the modal intensities. This capability will be critical in spectroscopic applications that often require GHz probing of low-pressure absorption features.

However, microwave injection locking often comes at the price of increased offset fluctuations and broader optical linewidth. Consequently, one should either fully stabilize the offset and repetition rate frequency, or leave both free-running. Passive full-FSR tuning assumes increasing importance in this context. Figure 7 shows the spectral properties of a device that resides in a sparse yet stable (narrow-linewidth) comb regime across a wide range of injection currents. At the highest currents, the spectrum becomes moderately
Figure 7. Passive current tuning over more than half the FSR. (a) Development of the optical spectrum measured with an FTIR. Although the intermode beat note remains narrow across the full scan range, only selected regimes (such as that in the framed panel) have a spectral structure sufficiently uniform for spectroscopy. (b) Evolution of the optical spectrum in a spectroscopically-compatible regime. Approximately 60% of the FSR can be covered with only minor changes in the modal intensity. (c) Simultaneously-measured microwave intermode beat note, which remains at a nearly constant position throughout the scan (previously-unpublished data).

sparse and hence spectroscopically useful. Some devices can passively tune by 60% of the full-FSR, which should improve significantly with future dispersion management techniques that will facilitate the development of combs operating over wider current and temperature ranges.

5. Modal sparsity and harmonic states

5.1. Harmonic states

Intracavity nonlinearities (typically $\chi^{(3)}$), along with GVD effects can induce a comb to operate in harmonic states. Although their origin in the novel ICL combs is poorly understood at present, we can only assume similarity to the related phenomena observed previously in QCL combs [63]. Instead of an intriguing anomaly, quasi-harmonic (sparse) states have in fact been observed in a majority of the ICL combs fabricated by the authors, and can produce broadband, low-noise comb operation.

Figure 8 plots FTIR interferogram (electric field autocorrelation) traces of various harmonic states (left), along with the corresponding optical spectra (right). Figure 8(a) shows the most desired fundamental comb state, which has a quasi-flat spectral envelope. The interferogram has clear bursts at intervals of $\Delta L = 3.09$ cm, which are determined by the comb’s repetition rate and separated by silent intervals. This state is characterized by a narrow (sub-kHz) intermode beat note, as shown above in figure 2(b).

Unfortunately, most ICL combs tend to operate in a quasi-harmonic (sparse) state like that plotted in figure 8(b), in which severe spectral modulation causes additional features to appear at multiples of $\Delta L/2$.

In fact, this state is not purely harmonic since modes spaced by the fundamental can still be seen in the spectrum. Furthermore, the periodicity of the interferogram remains unchanged. Under certain bias conditions, this sparse state may evolve into a pure 2nd order harmonic state such as that illustrated in figure 8(c). In this case, changing the injection current by only a few mA suppresses every other mode. The interferogram now has twice as many periodic bursts, whereas the spectrum has a slightly smoother envelope. The next subsection will analyze this transition more closely in the RF domain. Finally, ICL combs can support harmonic operation of extremely high order. Figure 8(d) shows lasing on every 12th mode (~3.87 cm$^{-1}$ spacing, 116.1 GHz), for which the interferogram bursts almost overlap. Such states may be useful for low-power submillimeter wave generation, or for liquid and solid spectroscopy when resolution of order 100 GHz is sufficient. Cavity asymmetries imposed by facet coatings [63] or defects can engineer this state to occur.

High-order harmonic states are more likely to arise in devices that initially display modal sparsity. Figure 9 illustrates a transition from sparse to high-order harmonic (12 cm$^{-1}$ spacing, 360 GHz), where the harmonic modes are somewhat more pronounced in the fundamental sparse state. Extreme mode spacings in the submillimeter wave range may be used to generate low-power terahertz waves, by relying on the ICL’s second order nonlinearity [50] or using mid-IR photoconductive antennas [64].
5.2. Harmonic states in the frequency domain

It is also useful to analyze the harmonic states in the RF domain, since mode skipping does not necessarily imply the preservation of a fixed phase relationship. The simplest (single-device) experiment compares the electrical intermode beat notes at the fundamental \( f_{\text{rep}} \) and second harmonic \( 2f_{\text{rep}} \) of the repetition rate, as shown in figure 10. While both beat notes exist in the sparse state, the second harmonic is several dB stronger despite considerable microwave losses at the higher frequency. The fundamental vanishes completely in the pure 2nd-order harmonic state, even though this may not be obvious from visual inspection of the frequency spectrum. The finite spectrometer resolution, along with Fourier spectrum side lobes, may give the impression that the fundamental lines are suppressed by only \( \sim 15 \) dB.

It is also useful to investigate how the transition between harmonic and sparse states affects dual-comb beating [65]. Figure 11 illustrates the beating of a local oscillator comb (LO, upper) with a signal comb (SIG, lower) whose state can be altered via injection current. Switching the SIG comb allows us to compare two cases: (a) one comb residing in the harmonic state, and (b) both in sparse states. Even in the harmonic state, all lines are mutually locked in compliance with the frequency comb model. The large mode spacing \( \sim 116 \) GHz, combined with an extremely high spectral refresh rate of the dual-comb signal \( 1/\Delta f_{\text{rep}} = 2.9 \) ns, potentially makes this dual-comb arrangement quite attractive for studying the chemical reaction kinetics of species with broadband absorption features, such as many hydrocarbons, with
Figure 9. Transition from sparse to high-order (40th) harmonic comb state at an injection current around 4.5 $I_{th}$. The separation between the harmonic comb lines reaches $\sim$360 GHz (previously-unpublished data).

Figure 10. Quasi-harmonic (sparse) and 2nd-order harmonic comb states in a 4 mm long ICL OFC. A change of injection current by 5 mA induced the transition to the harmonic state. In the quasi-harmonic state (top), the second harmonic of the repetition rate $2f_{rep}$ is stronger than the fundamental (even ignoring higher microwave losses at the higher frequency). While the optical spectrum is somewhat sparse, modes spaced by the fundamental frequency $f_{rep}$ are still present as indicated in the microwave spectrum. In contrast the 2nd-order harmonic state (bottom) skips every other mode, with more than 40 dB suppression of the fundamental. The darker structure in the optical spectrum stems merely from insufficient resolution and side lobes of the FTIR’s Fourier transform (previously-unpublished data).

unprecedented speed. For example, in microfluidic experiments a harmonic dual-ICL-comb spectrometer covering $\sim$0.5 THz of bandwidth may provide a valuable real-time MWIR probe [66]. On the other hand, the dual-comb spectrum is much denser when both combs are operated in sparse quasi-harmonic states. Further research is needed to probe the susceptibility of harmonic states to delayed optical feedback [63]. It was observed that poor alignment of the collimating lens for a QCL harmonic comb could preclude harmonicity, or cause the comb to enter a dense state similar to the sparse ones observed here. The same study showed the device could operate in completely different regimes, depending on the optical feedback associated with either diverging emission from the laser facet or collimation by an off-axis parabolic mirror or lens.
All of the ICL devices discussed here were collimated using a short-focal-length antireflective-coated lens that inevitably introduced some optical feedback. Given that the ICL combs in a recent frequency-stabilization experiment showed significant susceptibility to light returning to the cavity [67], we expect that even residual feedback may significantly impact the harmonic states as well. When an optical isolator was introduced into a recent multi-heterodyne experiment, the millisecond-scale optical linewidth narrowed by over an order of magnitude, from $\sim$5 MHz [41] to $\sim$350 kHz [42]. This confirms that even residual feedback from a downstream optical component can induce significant fluctuations of the offset and repetition rate.

6. Spectroscopy using ICL combs

The expanding interest in developing and optimizing OFCs is driven largely by the promise of portable, battery-operated, broadband high-resolution dual-comb spectrometers [68]. The optical down-conversion process in such a DCS takes place in a square-law photodetector, which produces an array of discrete microwave lines at frequencies separated by the difference in repetition rate $\Delta f_{\text{rep}}$ and offset by the difference in offset frequency $\Delta f_0$. Unique advantages include the absence of any moving parts and the ease of signal digitization, as well as the potential for full on-chip integration [34, 69, 70]. ICLs provide an especially flexible platform for the latter, since their bifunctionality allows sources and detectors to be processed from the same wafer material [55]. Clearly, a thermoelectrically-cooled HgCdTe photodetector, even given the frequency response required for DCS, is incompatible with this vision. Conventional photolithography can potentially be used to define a chip-scale mid-IR DCS photonic integrated circuit, which combines the two sources, detector, sensing waveguide, and passive connecting waveguides on the same chip [69, 70].

To date, the only demonstrations of DCS with ICL comb sources have been reported by the NRL/JPL team, sometimes in collaboration with Princeton University. The first experiments, at $\lambda \approx 3.2 \mu$m in 2017 [24], attained rapid (20 $\mu$s), broadband sensing of pure methane (CH$_4$) at atmospheric pressure, as well as swept high-resolution absorption and dispersion sensing of low-pressure ethylene at 12 Torr (1.6 kPa). Subsequently, methane (CH$_4$) and hydrogen chloride (HCl) were detected with much higher sensitivities at $\lambda \approx 3.6 \mu$m [41]. By increasing the effective path length to 76 m in a multi-pass Herriott cell, the limit for detecting HCl in 1 s was reduced to 100 ppb. The most recent DCS demonstration, in 2020, employed a self-contained, all-room-temperature apparatus (figure 12) that combined the ICL combs with an ICD fabricated from the same wafer [42]. The sensitivity for detecting difluoroethane, a broadband species, was nearly the same as in prior experiments despite room temperature operation of the ICD. The noise-equivalent absorbance (NEA) in all of these DCS demonstrations, quantified as the standard deviation (precision) at a given integration time, was of order $10^{-7}$–$10^{-4}$ $\sqrt{\text{Hz}^{-1}}$. This is similar to typical levels for direct absorption spectrometers that incorporate tunable single-mode lasers. A long optical interaction
length is prerequisite for ultra-high-sensitivity, while the NEA can be improved by exploiting wavelength modulation [59] or cavity ringdown.

Recent advances in computational phase correction [71,72] make it possible to perform DCS with completely free-running ICLs [41]. Although frequency-locked loops were initially required to stabilize the lasers, digital phase correction permits spectroscopy even in regimes with considerable phase noise [24], and eventually complete elimination of the loop [41]. In this approach, the free-running dual-comb beating signal, recorded with a fast digitizer, is post-corrected to retrieve the acquisition-time-limited linewidth by extracting the correction parameters from the signal itself. After adaptive resampling [73] and offset phase-shifting, the time-domain comb signal becomes phase-stable and useful for spectroscopic assessment. While these techniques still require considerable computational power, recent improvements in the feedback management will enable free-running ICL DCS to operate without any phase correction at all [42]. Finally, the high speed of digitizers is no longer a prerequisite when one employs techniques to sample the electrical signal below the Nyquist rate [74].

Note also that DCS is not the only spectroscopic technique compatible with ICL combs. Ongoing efforts are developing direct OFC techniques [75] that exploit the large mode spacing and optical cavity enhancement. Well-established Fourier spectroscopy can also benefit from an ICL comb as a bright, coherent source with equidistant lines [76, 77]. For convenience, figure 13 plots absorption cross-section spectra for some environmentally-important molecular targets (at atmospheric pressure and \(T = 298.1\) K) that are detectable with ICL combs, from the HITRAN 2012 [78] and HITRAN 2016 [79, 80] databases. Whereas sharp-line molecules like HCl are easily probed with single-mode lasers, comb spectroscopy is particularly advantageous for characterizing gas mixtures (multi-species sensing), isotope mixtures, or molecules with broad absorption features. Examples include longer-chain hydrocarbons like propane and butane, as well as natural gas leaks that comprise a mixture of alkanes.

Figure 12. Self-contained dual-comb spectrometer. (a) Schematic of the experimental setup; (b) optical spectra of the two combs; (c) power-averaged, free running dual-comb spectrum acquired in 2 ms; (d) experimental (points) and fit (curve) dual-comb spectrum for difluoroethane (refrigerant). Reprinted from [42], with the permission of AIP Publishing.

Figure 13. HITRAN 2012 [78] and HITRAN 2016 [79, 80] spectra of potential molecular targets for ICL comb spectroscopy. (a) Formaldehyde (H\(_2\)CO, green), nitrogen dioxide (NO\(_2\), dark brown), hydrogen chloride (HCl, light brown), and ozone (O\(_3\), blue). (b) Alkanes: methane (CH\(_4\), red), ethane (C\(_2\)H\(_6\), blue), propane (C\(_3\)H\(_8\), green), and butane (C\(_4\)H\(_10\), brown).
Figure 14. Spectral coverages for the available on-chip (fully integrated) frequency comb platforms. ICL data are taken from table 1, while the QCL and microresonator bandwidths follow [5]. The coverages for quantum dot (QDL), quantum dash (QDh), and quantum well diode laser (QWDL) frequency comb are from [33, 82]. Note that among the QDh/QDL/QWDL families, only the latter has demonstrated DCS.

7. Comparison with other on-chip platforms

Although numerous techniques can be used to generate OFCs, this review has focused on MWIR comb emission from fully-integrated chip-scale sources. It is of practical relevance to view the ICL comb platform in a broader perspective, which compares its spectral coverage with that of other small-footprint sources: QCLs, microresonators with integrated optical pumps, and conventional semiconductor lasers (which were not included in prior reviews). It was recently discovered that lasers based on QW, QD, and QDh gain media can act as stable, self-starting combs sources that exploit physical phenomena similar to those of QCLs or ICLs, and with nearly identical emission characteristics. Recent microwave interferometry characterization experiments on a QDL [81], and the use of QWDLs in DCS [33, 82], fully justify the inclusion of these lasers among novel monolithic comb sources.

Figure 14 graphically summarizes the different platforms, most of which operate in the NIR band of the electromagnetic spectrum (λ < 2 μm). While the repetition rates for most of the sources considered here fall in the 6–40 GHz range, microresonator-based sources can offer line spacings exceeding 100 GHz, or even multiples of that in harmonic states. Spectrally, only two sources (ICLs and microresonators) offer emission near 3 μm. Furthermore, the spectral window 3.5–4 μm, which is exceedingly important for sensing organic molecules, is unique to ICLs. Currently only QCLs can cover longer wavelengths extending from 4 μm to the LWIR and THz (60–70 μm and 100–200 μm) regions, although ICL combs operating in the 4–6 μm band should ultimately be quite feasible.

The typical values for instantaneous optical coverage are of order 1–3 THz for semiconductor-laser-based combs and ~50 THz for microresonators. Unfortunately, current ICL combs develop bandwidths on the lower side of this range, due to modal leakage that induces a periodic modulation of the laser gain and dispersion [48].

8. Conclusions and outlook

The preceding sections have outlined the brief history of ICL frequency combs and their capabilities to date. The unique, electrically-pumped, chip-scale ICL comb platform offers coherent, high-brightness MWIR emission with terahertz optical bandwidths and gigahertz repetition rates. Furthermore, the ICL’s bi-functionality feature may potentially allow dual-comb spectrometers to be fully-integrated on a chip that operates at room temperature with power consumption <1 W. The latter may be particularly useful in portable, battery-operated sensors of multiple species, or of chemicals with broad absorption features such as complex organic molecules.

Present ICL combs are still research-grade sources that need several key improvements before commercial utility can be claimed. While their compactness and output of 10 s of mW from a drive power budget <1 W are ideal for portable systems, the spectral properties of the low-noise states in most current ICL combs remain impractical for many sensing applications. This is due to the sparse spectra and occasional switching between different spectral patterns. Mode grouping, arising most likely from mode leakage into the high-index GaSb substrate, limits the maximum comb spectrum to a bandwidth well below that natively supported by the gain medium. On the other hand, devices emitting broader and more uniform spectra tend to display significant phase noise that precludes their application in DCS. It is therefore critical that
techniques to control the waveguiding and dispersion properties be developed, to provide routine ICL comb operation with high spectral uniformity and low phase noise.

Optimization of the ICL comb technology will also require further theoretical exploration of such issues as gain dynamics, the origin of harmonic states, and modal sparsity, as well as gain engineering to impose passive mode locking. It is also critical to minimize the sensitivity to optical feedback, which significantly affects the optical linewidth and triggers chaotic switching between comb regimes. Also needed is a deeper understanding of intensity and frequency noise mechanisms. Prior research that studied the linewidth broadening factor and frequency noise power spectral density in single-mode ICLs was reviewed in [18]. It was concluded that the intrinsic linewidth of \( \sim 10 \text{ kHz} \) ranks these sources between QCLs (with 100's of Hz intrinsic linewidth) and bipolar lasers [83]. Nevertheless, a multiplexed measurement performed on an ICL comb source should also be made as in [84], to characterize the frequency noise correlation between all of its lines. To date, multiheterodyne experiments on the best-performing ICL comb devices at 1 ms timescale have observed optical linewidths as low as 400 kHz, which is only slightly higher than for narrow-linewidth single-mode ICLs (\( \sim 300 \text{ kHz} \)) [85]. Unfortunately, this linewidth gradually increases across the comb spectrum due to timing jitter, which modulates the line spacing and yields accumulated frequency shifts for modes lying at the edges of the optical spectrum. Consequently, it is of key importance to understand the different mechanisms influencing the repetition rate stability, and in particular the technical noise. Regarding the relative intensity noise (RIN) characteristics, recent experiments by Deng et al performed on a Fabry–Pérot ICL [86] revealed lower RIN level with increasing injection current, but only in a single-mode and single-spectral lobe regime. Upon splitting into two spectral lobes, the RIN suddenly increased in the range 0–100 MHz. This observation is consistent with our early studies of ICLs used for multiheterodyne spectroscopy [23], where the appearance of a second spectral lobe was accompanied by a drastic increase of the RIN. Therefore, we anticipate that in comb regimes the RIN should not differ significantly from that of a single-mode laser, while in regimes with multiple spectral lobes possessing different GVDs, pronounced RF noise is expected. Of course, this prediction will require further experimental validation.

Ongoing development should also extend the wavelength coverage of ICL combs, e.g. to \( \lambda = 4–6 \text{ m} \), since power consumption will be much lower than for QCL combs and passive mode-locking is feasible. Another promising avenue is the use of optical nonlinearities to realize OFCs operating in multiple spectral bands simultaneously [50], or even to generate room-temperature terahertz combs through intracavity difference frequency generation [87]. ICL combs will also benefit from optical referencing to state-of-the-art metrological grade OFCs operating at telecom wavelengths. It should be possible to realize this by exploiting intracavity sum frequency generation to coherently up-convert the mid-infrared to the NIR, as we have already demonstrated at 3.6 \( \text{ m} \) [50].

**Data availability statement**

The data that support the findings of this study are available upon reasonable request from the authors.

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**ORCID iD**

Łukasz A Sterczewski  [https://orcid.org/0000-0003-1459-7517](https://orcid.org/0000-0003-1459-7517)

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