

Recent progress on mid-infrared and terahertz dual-comb spectroscopy

Zhuoren Wan, Min Li*, Binbin Liu, Yu Xia, Yuan Chen, Xiuxiu Zhang, Dongxu Zhu, Yan Dai, Sinan Tao, Hua Li*, Ming Yan* and Heping Zeng*

Citation: Wan ZR, Li M, Liu BB et al. Recent progress on mid-infrared and terahertz dual-comb spectroscopy. *Opto-Electron Technol* **2**, 260015 (2026).

<https://doi.org/10.29026/oet.2026.260015>

Received: 15 April 2026; Accepted: 21 July 2026; Published online: 22 September 2026

Related articles

Multistable soliton dynamics in an optical microresonator

Zichun Liao, Yuchong Cai, Lun Li, et al

Opto-Electronic Advances 2026, **9**(5): 250329 doi: [10.29026/oea.2026.250329](https://doi.org/10.29026/oea.2026.250329)

Soliton microcombs in optical microresonators with perfect spectral envelopes

Mulong Liu, Ziqi Wei, Haotong Zhu, et al

Opto-Electronic Advances 2025, **8**(3): 240257 doi: [10.29026/oea.2025.240257](https://doi.org/10.29026/oea.2025.240257)

Soliton microcomb generation by cavity polygon modes

Botao Fu, Renhong Gao, Ni Yao, et al

Opto-Electronic Advances 2024, **7**(8): 240061 doi: [10.29026/oea.2024.240061](https://doi.org/10.29026/oea.2024.240061)

Terahertz imaging technology: progress and applications

Yuyuan Tian, Xiaoyin Chen, Zhuocheng Zhang, et al

Opto-Electronic Technology 2026, **2**(1): 250009 doi: [10.29026/oet.2026.250009](https://doi.org/10.29026/oet.2026.250009)

More related articles in Opto-Electronic Journals Group website



Recent progress on mid-infrared and terahertz dual-comb spectroscopy

Zhuoren Wan¹, Min Li^{2*}, Binbin Liu^{3,4}, Yu Xia¹, Yuan Chen¹, Xiuxiu Zhang¹, Dongxu Zhu¹, Yan Dai¹, Sinan Tao², Hua Li^{3,5*}, Ming Yan^{1,5*} and Heping Zeng^{1,4,6*}

Abstract: Dual-comb spectroscopy (DCS) has emerged as a revolutionary optical technique, offering exceptional spectral resolution, rapid measurement capability, and inherent frequency accuracy without the need for moving parts. This review provides a comprehensive survey of the principles and technological developments of DCS within the mid-infrared (mid-IR, $\sim 2\text{--}20\ \mu\text{m}$) and terahertz (THz, $\sim 0.1\text{--}10\ \text{THz}$) spectral regions. These regimes are of paramount importance due to the unique molecular fingerprints and low-energy excitations they encompass, spanning areas such as gas-phase chemical analysis, combustion diagnostics, and atmospheric sensing. We systematically outline the primary approaches for generating dual-comb sources in these spectral ranges, including direct generation via quantum cascade lasers, optical parametric oscillation, difference frequency generation, and microresonator-based frequency combs. The performance metrics, trade-offs, and recent innovations for each platform are critically compared. Finally, we discuss key challenges and emerging directions, including continuous mid-infrared-to-THz spectral coverage, broadband and low-noise detection, beam quality and mode matching, quantum-enhanced sensitivity, and coherence preservation for long-term operation. We conclude by outlining future trends toward integrated, field-deployable, and application-oriented DCS systems capable of broadband molecular fingerprinting, high-speed sensing, and precision spectroscopy.

Keywords: optical frequency combs; dual-comb spectroscopy; mid-infrared spectroscopy; terahertz spectroscopy

DOI: [10.29026/oet.2026.260015](https://doi.org/10.29026/oet.2026.260015) | CSTR: [32419.14.oet.2026.260015](https://cstr.cn/32419.14/oet.2026.260015)

Citation: Wan ZR, Li M, Liu BB et al. Recent progress on mid-infrared and terahertz dual-comb spectroscopy. *Opto-Electron Technol* **2**, 260015 (2026).

1 Introduction

Optical frequency combs (OFCs) consist of evenly spaced coherent spectral lines, offering broad spectral coverage, high resolution, and absolute frequency traceability. They have been widely applied in precision metrology^{1–3}, molecular fingerprinting^{4,5}, and atmospheric and environmental monitoring⁶. For instance, recent studies have demonstrated the use of vacuum-ultraviolet OFC to directly excite the $^{229\text{m}}\text{Th}$ nuclear clock transition, establishing a frequency link to existing atomic clocks and further highlighting the

transformative role of OFCs in precision metrology⁷. Meanwhile, OFCs have enabled significant advances in molecular fingerprinting by providing rapid and accurate spectral identification of complex gas mixtures. For example, broadband direct comb spectroscopy operating in the mid-infrared region has been used to simultaneously monitor multiple trace greenhouse gases—such as methane, carbon dioxide, and nitrous oxide—with part-per-billion sensitivity in both laboratory and field settings⁸. In atmospheric and environmental monitoring, portable optical frequency comb systems have been deployed for real-time detection of

Received: 15 April 2026

Accepted: 21 July 2026

Published online: 22 September 2026

¹State Key Laboratory of Precision Spectroscopy, and Hainan Institute, East China Normal University, Shanghai 200062, China; ²School of Optical Electrical and Computer Engineering, University of Shanghai for Science and Technology, Shanghai 200093, China; ³Key Laboratory of Terahertz Solid State Technology Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, Shanghai 200050, China; ⁴Chongqing Key Laboratory of Precision Optics, Chongqing Institute of East China Normal University, Chongqing 401121, China; ⁵Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China; ⁶Jinan Institute of Quantum Technology, Jinan 250101, China.

*Correspondence: M Li, E-mail: minli1220@usst.edu.cn; H Li, E-mail: hua.li@mail.sim.ac.cn;

M Yan, E-mail: myan@lps.ecnu.edu.cn; HP Zeng, E-mail: hpzeng@phy.ecnu.edu.cn

atmospheric pollutants, including volatile organic compounds and nitrogen oxides, supporting urban air quality assessment and emission source tracking⁹. These applications underscore the versatility of OFC technology in addressing pressing scientific and societal challenges across different spectral domains and measurement environments.

Among the OFC-enabled spectroscopic techniques, dual-comb spectroscopy (DCS) stands out as one of the most powerful tools for time-resolved, high-resolution measurements. Derived from the interferometric beating of two coherent OFCs, DCS leverages multi-heterodyne detection to map optical spectra into the radio-frequency (RF) domain, thereby retaining the absolute frequency accuracy of the original combs^{10–13}. By operating without moving parts, DCS overcomes key limitations of conventional spectrometers—such as restricted acquisition speed, limited spectral resolution, and compromised frequency accuracy—associated with traditional dispersive instruments^{4,14} and Michelson-interferometer-based Fourier-transform spectroscopy (FTS)^{15,16}. For instance, when combined with Doppler-free techniques¹⁷, DCS enables sub-Doppler spectroscopy with frequency resolutions reaching the megahertz (MHz) level¹⁸. In terms of measurement speed, DCS exploits the small repetition rate difference (Δf_r) between the two combs (typically ranging from 100 Hz to 1 MHz) to enable asynchronous sampling without mechanical scanning, yielding a minimum acquisition time ($1/\Delta f_r$) down to the microsecond (μ s) and nanosecond (ns) regimes¹³. Recent advances—including chip-scale DCS^{19,20} with sub-GHz Δf_r and programmable combs combined with compressive sensing²¹—have further pushed DCS toward real-time operation. With respect to spectral bandwidth, nonlinear frequency-conversion techniques^{22,23} have enabled OFCs broadening spanning up to six octaves²⁴, offering a direct pathway to extend the single-shot spectral coverage of DCS. In addition, the development of ultraviolet photon-counting DCS²⁵ and THz DCS²⁶ has further expanded the accessible measurement range. From a sensitivity perspective, DCS is compatible with a wide range of enhancement techniques, including optical cavities²⁷, multipass cells²⁸, hollow-core fibers²⁹, and surface plasmon resonance³⁰, enabling gas detection sensitivities at the part per million (ppm) level. To overcome limitations imposed by detector background noise, DCS has also been combined with photo-acoustic³¹, photo-thermal³², and optomechanical³³ spectroscopy, pushing detection sensitivities into the part per billion (ppb) regimes.

While near-infrared DCS has demonstrated outstanding performance³⁴, recent research efforts have increasingly shifted toward the mid-infrared and terahertz (THz) spectral regions. This shift is driven by the fundamental nature of spectroscopy: the mid-infrared encompasses the molecular fingerprint region, where fundamental vibrational transitions are several orders of magnitude stronger than near-infrared overtone and combination bands, enabling

enhanced absorption contrast, sensitivity, and quantitative molecular identification³⁵. The THz region, meanwhile, provides access to molecular rotational transitions, weakly bound states, and low-frequency collective modes in solids, offering unique insights into intermolecular interactions and condensed-matter dynamics³⁶. Extending DCS into the mid-infrared and THz therefore enables broadband and ultrasensitive molecular fingerprint analysis, substantially broadening its application scope in trace-gas sensing, reaction dynamics, biomedical diagnostics, and precision spectroscopy. The rapid advancement of mid-infrared and THz DCS calls for a comprehensive review that synthesizes recent breakthroughs, clarifies technical challenges, and identifies emerging application pathways. Unlike prior reviews that typically focus on specific source platforms or spectral regions, this review provides a unified perspective on recent advances in mid-infrared and THz DCS. Beyond summarizing representative demonstrations, we emphasize recent developments in source architectures, coherence-preservation strategies, and the associated performance trade-offs. We also discuss key cross-band challenges, including spectral coverage, detection sensitivity, system complexity, long-term stability, and field deployability. By comparing the mid-infrared and THz regions side by side, this review highlights both shared methodological bottlenecks and complementary application opportunities, with the aim of offering an accessible entry point and a perspective on future directions in spectroscopic development.

This article is organized as follows. Section 2 provides a brief overview of DCS, emphasizing its basic scaling laws. Section 3 surveys generation techniques and recent progress in mid-infrared DCS. Section 4 reviews generation techniques and advances in THz DCS. Section 5 surveys the key challenges and emerging directions in DCS. Section 6 concludes with an integrated synthesis of recent advances and future perspectives.

2 Basic principle

Figure 1(a) shows the classic asymmetric DCS scheme for high-precision spectral analysis. When the comb pulses propagate through a medium, the spectral information is encoded in the amplitude and phase of the optical pulse sequence and comb modes. By using linear optical sampling techniques, optical interferograms can be obtained, from which the encoded spectral information can be decoded. Figure 1(b) illustrates the time-domain and frequency-domain principles of the DCS linear sampling process. Practical measurements normalize out comb-to-comb spectral structure using a reference path or interleaved backgrounds.

In the time domain, DCS consists of two pulse sequences with slightly different repetition rate (f_{r1} , $f_{r2} = f_{r1} + \Delta f_r$). One pulse sequence scans the other with a step size of $\Delta T = \frac{1}{f_r} - \frac{1}{f_r + \Delta f_r} \approx \frac{\Delta f_r}{f_r^2}$, generating a time-domain

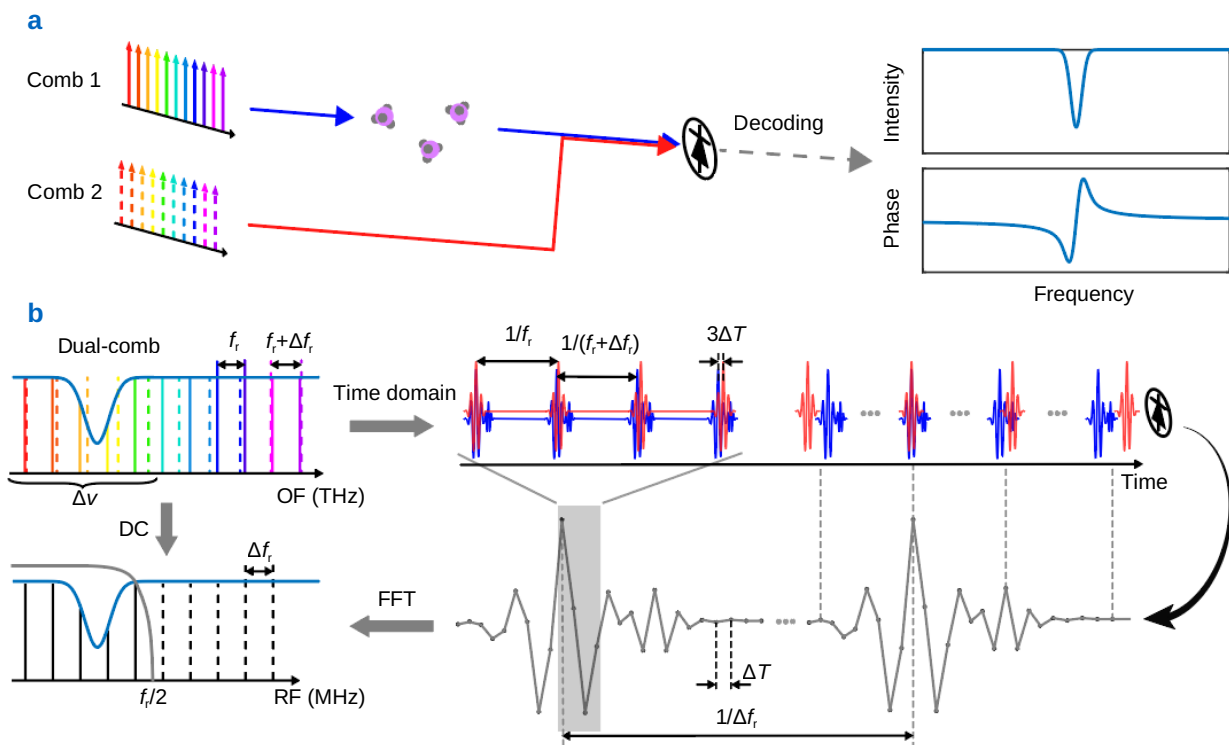


Fig. 1 | Basic principle of DCS. (a) System architecture. (b) Time and frequency domain characteristics of DCS.

interference signal. After being detected by a photodetector, the interference signal is reproduced with a period of $1/\Delta f_r$, effectively down-sampling the optical signal.

In the frequency domain, each pair of comb teeth generates a frequency-downshifted RF beat signal, forming an RF comb with a spacing of Δf_r . A Fourier transform of the time-domain interferogram results in the RF comb, where the down-conversion mechanism shifts an optical spectrum with bandwidth $\Delta\nu$ to an RF spectrum with bandwidth $\Delta\nu/m$, where $m = f_r/\Delta f_r$ is the conversion factor, typically ranging from 10^4 to 10^6 . This allows the optical frequency (OF) domain covering hundreds of THz, containing hundreds of thousands of comb teeth, to be mapped to a detectable RF range within the hundreds of MHz. To avoid spectral aliasing, all optical spectral information is mapped into the RF bandwidth range $(f_r/2)$, with the corresponding optical bandwidth being $\Delta\nu \leq \frac{mf_r}{2} = \frac{f_r^2}{2\Delta f_r}$. This relation highlights an important trade-off: increasing Δf_r shortens the interferogram period and improves the spectral refresh rate, but it reduces the alias-free optical bandwidth. Conversely, reducing Δf_r allows broader optical coverage, but increases the acquisition time for a single interferogram and places stronger demands on mutual coherence during averaging. Detector performance also imposes an important practical constraint on DCS. The detector must support the down-converted RF bandwidth and must provide sufficient linearity, dynamic range, and sensitivity. In the mid-infrared, high-speed detectors often require cooling and

involve trade-offs among responsivity, noise, saturation power, and bandwidth; these limitations become more severe toward the long-wave mid-infrared. In the THz region, detector choices are even more limited, and photoconductive, electro-optic, or self-mixing detection schemes each offer different compromises between bandwidth and sensitivity. These detector bottlenecks motivate several indirect or hybrid detection strategies discussed later, including up-conversion detection, electro-optic sampling, cross-comb spectroscopy, and self-detected quantum cascade laser (QCL)/THz-QCL dual-comb systems.

The minimum time required to resolve the RF comb teeth is $1/\Delta f_r$. However, this speed advantage is offset by the low signal-to-noise ratio (SNR) of a single spectral acquisition. For DCS with M comb teeth, the SNR at the shot-noise limit is $\sqrt{\tau n_{\text{comb}}}/M$, where τ is the sampling time, and n_{comb} is the number of photons detected per second. In wideband measurements, the SNR decreases with the increase in comb teeth (optical spectral bandwidth $\Delta\nu = Mf_r$), and in practical applications, multiple spectra must be co-added. Consequently, broadband measurements typically require coherent averaging of multiple spectra, with the minimum acquisition time ultimately determined by the target SNR. Such coherent averaging critically relies on maintaining mutual coherence between the two combs. It can be achieved by phase-locking both f_r and the carrier-envelope offset frequency (f_{ceo}) to a common reference or by real-time phase correction in software.

We further note that the repetition frequency f_r plays a

dual role. Increasing f_r enlarges the alias-free bandwidth for a given Δf_r and can support higher measurement speeds; however, it also increases the comb spacing, thereby reducing the spectral sampling density within a single acquisition. In contrast, a lower f_r provides denser spectral sampling and is advantageous for high-resolution gas-phase spectroscopy, but for a fixed optical bandwidth it increases the number of comb teeth, requiring a smaller Δf_r to avoid aliasing and placing greater demands on detection bandwidth and coherence stability. These trade-offs explain why broadband mid-infrared and THz DCS systems often require balancing spectral coverage, acquisition speed, spectral sampling density, detector bandwidth, and long-term mutual coherence. They also motivate different system-level strategies, including GHz-spacing combs for rapid acquisition, lower-repetition-rate combs for dense sampling, spectral filtering or stitching to extend bandwidth, and spectral interleaving to achieve effective resolution finer than the native comb spacing.

Moreover, an important consideration in DCS is the architecture used to generate two mutually coherent combs. The most common approach employs two independent mode-locked lasers referenced to a common optical or microwave standard, offering high flexibility but requiring active stabilization or post-processing correction of relative phase and timing noise. Alternatively, the two combs can be derived from a common seed laser or oscillator through electro-optic modulation. These common-source schemes suppress common-mode noise and simplify mutual stabilization, although they may suffer from limited spectral bandwidth or number of comb lines. A third approach is the single-cavity dual-comb architecture, where two pulse trains or modes share the same cavity, providing passive mutual coherence and reduced stabilization requirements. However, such systems often face challenges such as cross-talk between the single-cavity dual combs. This may arise from multiple factors such as shared gain media, residual coupling, nonlinear interactions, pump depletion, and environmental perturbations. Excessive cross-talk can generate parasitic RF signals, distort dual-comb spectra, reduce comb independence, and degrade quantitative retrieval performance. However, under certain conditions, controlled cross-talk can also be advantageous. For example, Fellingner et al. exploited the intrinsic intensity modulation present in a single-cavity dual-color dual-comb laser to extend the ambiguity-free ranging distance while preserving interferometric precision³⁷. Nevertheless, these architectures provide a useful framework for understanding the difference-frequency generation (DFG)-, optical parametric oscillator (OPO)-, microresonator-, QCL-, and THz-based dual-comb platforms discussed in the following sections.

3 Mid-infrared dual-comb spectroscopy

In the strategically important mid-infrared bands, DCS is still maturing compared to its well-established counterparts

in the visible and near-infrared ranges. Recent advances in nonlinear platforms—including DFG, microresonator frequency combs, and supercontinuum sources—have enabled the extension of DCS into the mid-infrared molecular fingerprint region. This progress provides a solid foundation for precise molecular identification and quantitative analysis. This section reviews the latest developments in mid-infrared DCS.

3.1 Difference-frequency generation (DFG)

DFG is a second-order nonlinear optical process in which two input waves at frequencies ω_1 and ω_2 mix within a $\chi^{(2)}$ medium to generate radiation at the idler frequency $\omega_{\text{idler}} = |\omega_1 - \omega_2|$ ³⁸. In practice, the accessible wavelength range is largely determined by the nonlinear medium: periodically poled lithium niobate (PPLN) is the workhorse platform for the 3–5 μm range owing to its high effective nonlinearity, mature fabrication technology, and flexible quasi-phase matching, whereas longer-wavelength materials such as GaS, GaSe, orientation-patterned GaP (OP-GaP), and orientation-patterned GaAs (OP-GaAs) enable comb generation in parts of the 5–20 μm region, where many heavier and more complex molecules exhibit strong fundamental vibrational fingerprints. A comparison of representative nonlinear crystals for DFG-based mid-infrared comb generation and DCS is summarized in Table 1.

Because the idler output directly inherits the coherence of the inputs, DFG has become one of the most widely used approaches for extending DCS into the mid-infrared region. For instance, DFG between a near-infrared comb and a continuous-wave (CW) laser has been widely used for generating mutually coherent mid-infrared comb. A representative example is the work of Baumann et al. (Fig. 2)³⁹, where a mid-infrared OFC centered near 3.4 μm was generated by DFG mixing an Er-doped fiber comb with a CW laser in a PPLN. All optical frequencies were referenced to a common ultra-stable cavity, resulting in an optical bandwidth of about 4.5 THz and gas measurements with SNR as high as 3500. Further extended spectral bandwidth (over 8 THz in a single-shot measurement) was achieved by using a specialized PPLN crystal⁴⁰. Meanwhile feed-forward stabilization of the relative f_{ceo} provided long-term coherence exceeding half an hour with a quality factor of $1.1 \times 10^6 \text{ s}^{-1/2}$. By adjusting the crystal temperature and polarization period, a measurement range of 82–100 THz could be achieved.

While traditional DCS relies on mode-locked lasers with complex phase-locking electronics, electro-optic (EO) frequency combs have emerged as a simpler and more agile alternative owing to their straightforward locking schemes and flexible frequency control⁴¹. Leveraging these advantages, EO-based DCS based on DFG have been demonstrated for mid-infrared spectroscopy (Fig. 3)⁴². The DFG process produces a flat-top mid-infrared DCS with tunable

Table 1 | Representative nonlinear crystals for mid-infrared DFG-based comb generation. The nonlinear figure of merit is defined as d^2/n^3 , with units of pm^2/V^2 . Here, d is the effective nonlinear coefficient and n denotes the refractive index.

Nonlinear process	Crystal	Nonlinear figure of merit (d^2/n^3)	Transparency range (nm)
Phase matching	CdSiP ₂	247	660–6500
	ZnGeP ₂	187	740–12000
	GaSe	143	650–18000
	AgGaSe ₂	61	710–18000
	BaGa ₄ Se ₇	29	470–18000
	AgGa ₂ S ₂	11	500–13000
Quasi-phase matching	OP-GaAs	245	900–17000
	OP-GaP	164	600–11000
	PPLN	56	330–5500
	PPKTP	38	350–4500
	PPKTA	37	350–4000

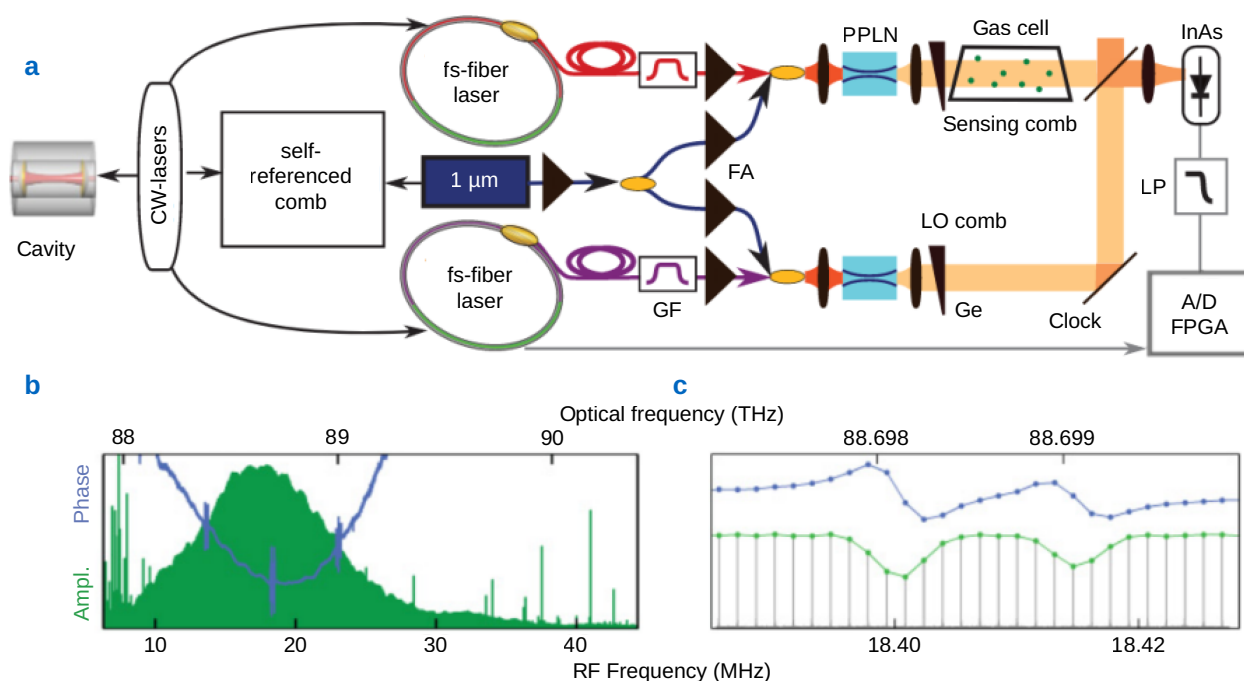


Fig. 2 | DFG-based DCS via PPLN. (a) Experimental setup and (b) Results with (c) enlarged view of the RF comb teeth. Figure reproduced with permission from ref.³⁹, American Physical Society.

central wavelength and repetition rate, enabling spectral measurements in the 3.15–3.5 μm range with Doppler-limited resolution in milliseconds. To expand the tuning range of the DCS, an EO-based DCS was generated in the near-infrared, with a tunable OPO as the pump, and the system used DFG to convert to mid-infrared region⁴³. The system covered the 3–4.7 μm mid-infrared region, with a comb repetition rate of 500 MHz, detecting approximately 440 comb modes and achieving an SNR of over 10^5 in one second. To increase the single-shot spectral bandwidth, cascading EO modulation combined with DFG has been employed to generate mid-infrared DCS centered near 3.3

μm , achieving a bandwidth of 442 GHz and a resolution of 50 MHz in a single measurement with a quality factor of $2.94 \times 10^6 \text{ Hz}^{1/2}$ in a 183.6 ms measurement time⁴⁴. Recently, EO-based DCS in the 8 μm region has been demonstrated for cavity-enhanced chemical analysis⁴⁵.

Despite their conceptual simplicity, comb-CW mixing schemes are fundamentally limited by the low peak power of the CW field, leading to weak nonlinear interaction and consequently reduced mid-infrared output power and optical SNR, especially beyond 5 μm ⁴⁶. To overcome these limitations, pulse-based DFG schemes have been widely explored. A signal frequency comb is mixed with a

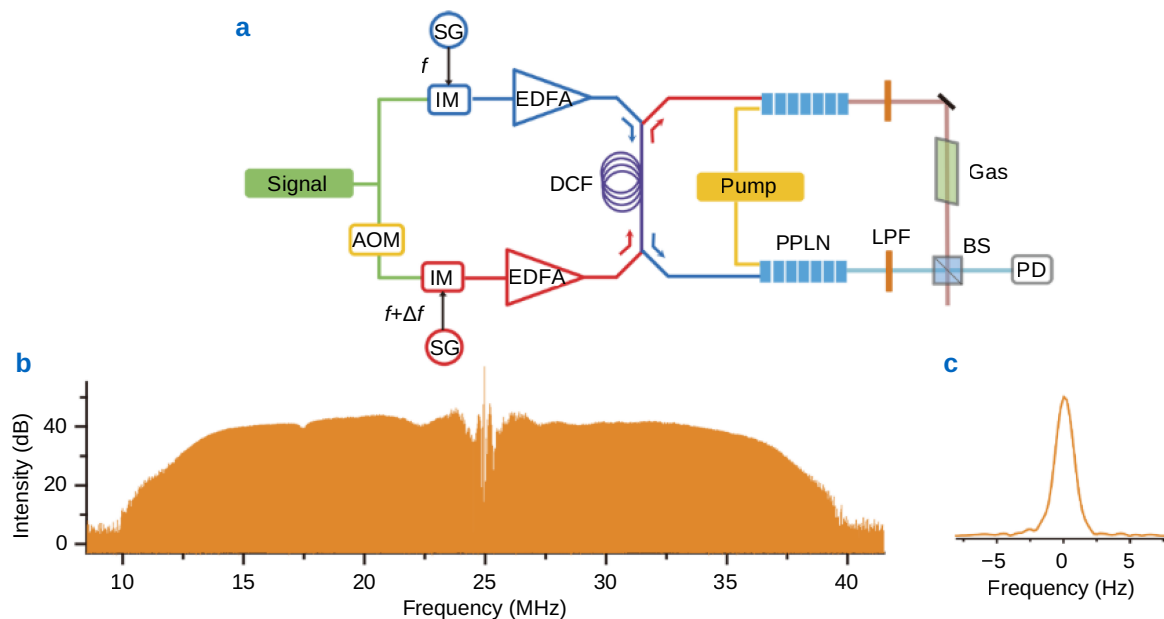


Fig. 3 | Schematic diagram of an EO modulated mid-infrared DCS based on DFG. (a) Layout of the system and (b) RF spectrum with (c) enlarged view of the RF comb teeth. Figure reproduced with permission from ref.⁴², Springer Nature.

femtosecond pump pulse in a nonlinear crystal such as PPLN, enabling substantially higher peak intensities and enhanced conversion efficiency. Using this approach, mid-infrared output powers of 40–100 milliwatt (mW) in the 2.5–3.8 μm range and 1–10 mW in the 4–5 μm range was demonstrated, sufficient to support open-path and long-path spectroscopic measurements⁴⁷. These schemes require precise control of the relative delay between the two pulse trains, making the system sensitive to mechanical and thermal drifts and limiting long-term stability.

To overcome these limitations, intra-pulse DFG (IP-DFG) mixes different spectral components within a single ultra-short pulse. This self-referenced process intrinsically preserves temporal overlap and phase coherence, eliminating the need for external delay stabilization while enabling broadband, high-power mid-infrared frequency comb generation. By using a commercially available 1.56 μm mode-locked laser, robust fiber amplification, and IP-DFG in various $\chi^{(2)}$ nonlinear crystals, DCS has been extended to cover spectral regions spanning approximately 3–13 μm (Fig. 4)⁴⁸. In a single-shot implementation, a broadband DCS (6.6–11.4 μm) was generated via IP-DFG in ZnGeP₂ crystals, driven by sub-three-cycle (~ 20 femtoseconds, fs) pulses at 2.4 μm ⁴⁹. The system achieved real-time acquisition rates of 0.1–12 seconds per spectrum, resolving over 240,000 comb modes with a spacing of approximately 80 MHz.

Table 2 compares various DFG-based mid-infrared DCS implementations, highlighting their performance trade-offs. In short, CW-pumped DFG emphasizes coherence and frequency accuracy but is limited by low mid-infrared power. EO-comb DFG offers compactness, electronic

tunability, and passive mutual coherence, whereas its bandwidth is often limited and requires tuning or stitching. Pulse-pumped DFG improves brightness in the 3–5 μm region but requires precise pulse synchronization and phase correction. IP-DFG provides offset-free broadband access to the long-wave mid-infrared, although it relies on few-cycle drivers and suitable nonlinear crystals. Thus, DFG remains one of the most flexible routes to mid-infrared DCS, while its practical performance is often constrained by conversion efficiency, phase matching, and long-term stabilization. Future advances in mid-infrared DCS are expected to benefit from wider transmission window crystals and higher-efficiency nonlinear processes, paving the way for broadband spectrum mid-infrared DCS with enhanced performance.

3.2 Mid-infrared DCS with OPOs

Beyond DFG-based approaches, OPOs constitute another powerful platform for mid-infrared DCS. In an OPO, a pump field drives parametric down-conversion in a nonlinear crystal, giving rise to mutually coherent signal and idler waves that simultaneously satisfy energy conservation and phase-matching conditions. When pumped by OFCs, the comb structure can be efficiently transferred to the mid-infrared. A major advantage of OPO-based DCS is their substantially higher output power and brightness, which translate directly into enhanced SNR and faster acquisition rates.

Early demonstrations of mid-infrared OPO-based DCS employed synchronously pumped OPOs to access the C-H stretching region near 3.3 μm , achieving approximately 172 nanometers (nm) bandwidth with GHz-level spectral

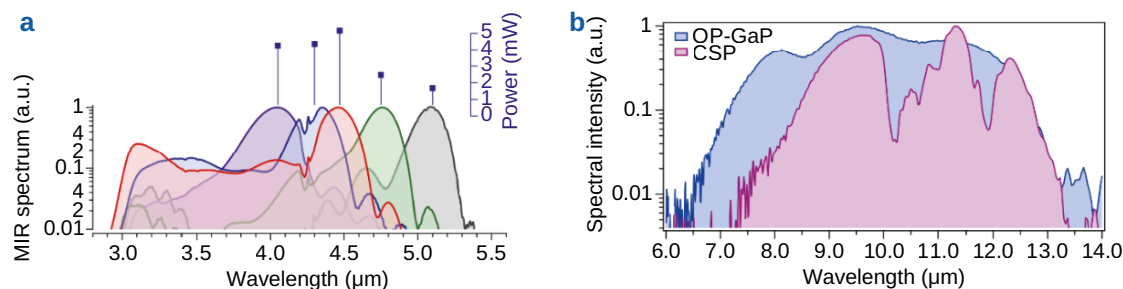


Fig. 4 | Broadband mid-infrared comb spectrum. (a) Spectrum from 3–5 μm generated using PPLN crystals with different poling periods. (b) Spectrum from 7–13 μm generated using CSP and OP-GaP crystals. Figure reproduced with permission from ref.⁴⁸, under a Creative Commons Attribution License.

Table 2 | Summary of representative DFG-based mid-infrared DCS architectures.

Architecture	Spectral coverage	Resolution/comb spacing	FOM	Output power	Main strengths	Main limitations
CW-pumped DFG with mode-locked combs ^{39,40}	82–100 THz tunability; 8.2 THz demonstrated span ⁴⁰	Typically ~100 MHz	$1.1 \times 10^6 \text{ Hz}^{1/2}$ ⁴⁰	Typically below 100 μW	High coherence and frequency accuracy	Low MIR power
CW-pumped DFG with EO combs ^{42–45}	Typically tunable across the 3–5 μm region, with extension to 7.77–8.22 μm ⁴⁵	Electronically tunable 100–500 MHz; 50 MHz reported ⁴⁴	$2.94 \times 10^6 \text{ Hz}^{1/2}$ ⁴⁴	Typically sub-mW to ~1 mW	Compact, electronically agile, and passively coherent	Limited instantaneous bandwidth
Pulse-pumped DFG ⁴⁷	Typically tunable across the 2.6–5 μm region	Typically ~100 MHz	the order of $10^6 \text{ Hz}^{1/2}$ ⁴⁴	Typically several mW to ~100 mW	High brightness and broad 3–5 μm coverage	Requires precise pulse synchronization
IP-DFG ^{48,49}	3–13 μm source coverage ⁴⁸ ; 6.6–11.4 μm ⁴⁹	Typically ~100 MHz; 1 GHz source platform ⁴⁸	$7.3 \times 10^6 \text{ Hz}^{1/2}$ ⁴⁹	mW-level ⁴⁸ ; up to 300 mW ⁴⁹	Offset-free generation with broad long-wave MIR access	Requires few-cycle drivers and specialized nonlinear crystals

resolution⁵⁰. Later implementations used pairs of ultra-broadband subharmonic OPOs pumped by phase-locked thulium-fiber combs. These systems achieved spectral coverage spanning approximately 3.1–5.5 μm , resolving more than 3.5×10^5 comb teeth with single-interferogram acquisition times as short as 7 ms. In addition, ppb-level detection sensitivity and inter-comb relative linewidth of 25 millihertz were demonstrated⁵¹.

In parallel, efforts have also been made to reduce system complexity by relaxing phase-locking requirements. A simpler architecture based on two free-running femtosecond OPOs has demonstrated that absolute optical precision can be recovered by simultaneously measuring f_r and f_{ceo} , without the need for optical phase locking (Fig. 5)⁵². Their system achieved a resolution of about 0.5 inverse centimeters (cm^{-1}) and an optical bandwidth greater than 60 nm for gas monitoring. Despite these advantages, the presence of an optical cavity inherently makes OPOs sensitive to environmental perturbations. As a result, stable OPO-based DCS operation typically requires complex active stabilization schemes, which increase system complexity and limit long-term stability.

To address these challenges, single-cavity OPO architectures have been proposed and experimentally demonstrated. By sharing a common cavity (Fig. 6), single-cavity OPOs intrinsically suppress relative noise between the two combs, enabling time-resolved DCS with a bandwidth of approximately 300 nm around 3.3 μm , a spectral resolution of 6 GHz, and a temporal resolution of 20 μs ⁵³. Recent advancements have further improved the practicality, stability, and output power of OPO-based DCS. For example, single-cavity, free-running dual-comb OPOs generating two idler combs near 3.5 μm have demonstrated output powers with 245 mW per branch and a relative intensity noise lower than -158 dBc/Hz ⁵⁴. More recently, the same research direction was extended in two important ways. First, a widely tunable free-running OPO combined with dual-comb and cross-comb detection enabled high-sensitivity spectroscopy across the infrared, with signal tuning from 1300 to 1670 nm and idler tuning from 2700 to 5000 nm⁵⁵. Second, dual-comb operation was pushed to a 1 GHz repetition rate in a femtosecond OPO, demonstrating that high repetition rates can be combined with the compactness and mutual-coherence advantages of shared-cavity architectures; this

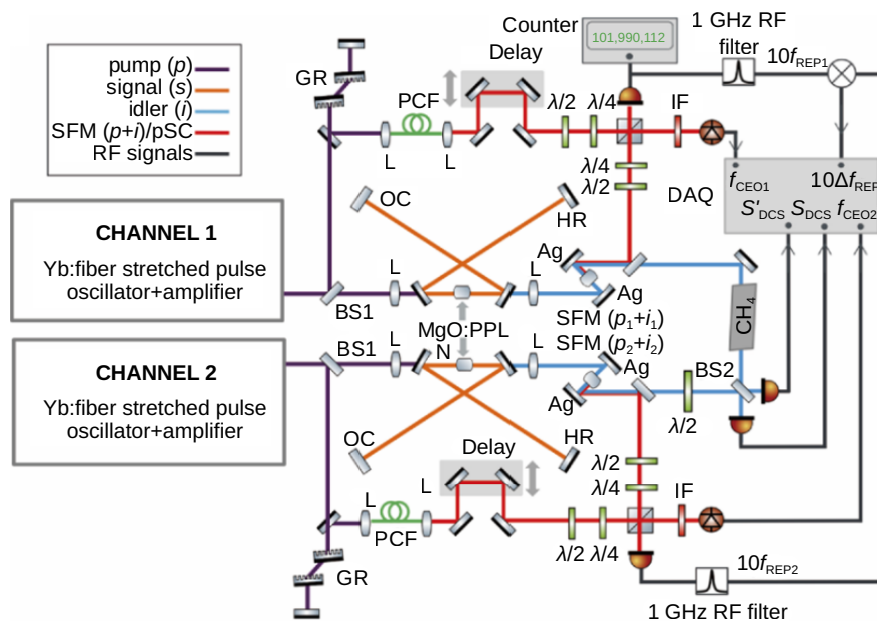


Fig. 5 | Free-running femtosecond OPOs. Figure reproduced with permission from ref.⁵², under the terms of the Optica Open Access Publishing Agreement.

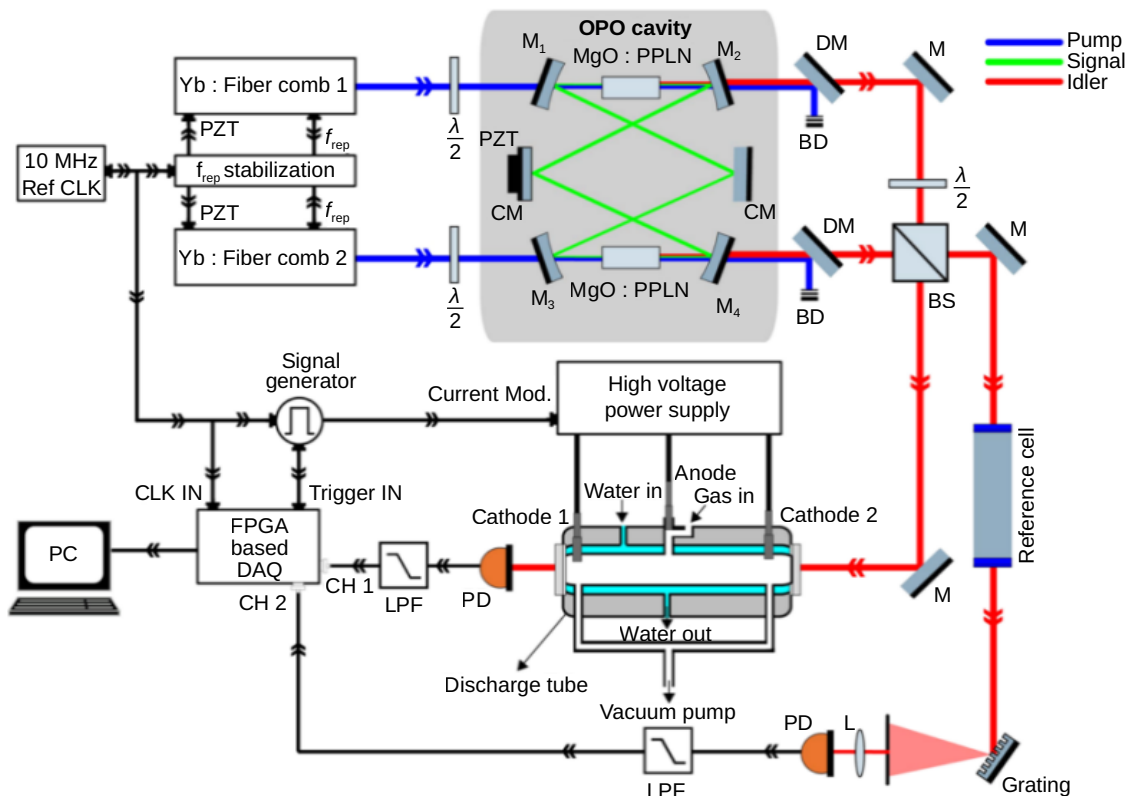


Fig. 6 | Single-cavity dual-crystal OPO scheme. Figure reproduced with permission from ref.⁵³, under a Creative Commons Attribution License.

platform also enabled 41 m open-path CH₄ measurements without active frequency locking⁵⁶.

To highlight the main architectural trade-offs, representative OPO-based mid-infrared DCS systems are compared in Table 3. OPO-based DCS has evolved from high-power

broadband implementations toward architectures with reduced stabilization complexity and improved practical sensitivity. Phase-locked subharmonic OPOs provide broad instantaneous coverage and excellent coherence, but require complex pump-comb and OPO-cavity stabilization.

Table 3 | Summary of representative OPO-based mid-infrared DCS architectures.

Architecture	Spectral coverage	Resolution	FOM	Output power	Main strengths	Main limitations
OPO-based high-power DCS ^{50,51}	3.1–5.5 μm	Typically tens MHz	$\sim 1.3 \times 10^6 \text{ Hz}^{1/2}$ ⁵¹	Adjusted from a few mW to $\sim 70 \text{ mW}$	High MIR brightness and broad instantaneous coverage	Complex pump-comb and OPO-cavity stabilization is required
Free-running OPO-DCS with frequency calibration ⁵²	Typically tunable across the 2.7–5 μm region	0.5 cm^{-1} resolution	FOM N.R. ⁵²	$\sim 100 \text{ mW}$	Simpler operation	Free-running fluctuations limit coherent averaging
Single-cavity / shared-cavity OPO architectures ^{53–56}	Typically tunable over 2.7–5.0 μm	80 MHz to 1.03 GHz comb spacing	$3.5 \times 10^8 \text{ Hz}^{1/2}$ ⁵⁵	200–320 mW	Suppress relative noise and support high per-line power	Cross-talk and reduced independent comb control must be considered

Free-running OPO-DCS simplifies operation through RF-based calibration or spectral registration, although coherent averaging and frequency accuracy remain limited by free-running fluctuations. Single-cavity and shared-cavity OPOs further suppress relative noise and support high per-line power, but careful management of cavity dispersion, spectral overlap, and comb-to-comb cross-talk is required. Owing to their excellent power scalability and spectral tunability, OPOs have emerged as a leading solution for high-performance mid-infrared DCS. Nevertheless, the wavelength coverage of OPO-based DCS is ultimately constrained by the transparency and phase-matching conditions of the nonlinear crystals employed, with most practical implementations operating predominantly in the 3–5 μm spectral region. Extending OPO-based DCS toward longer mid-infrared wavelengths often requires specialized materials and increasingly complex cavity designs, which can further limit system robustness and scalability. In contrast, QCLs inherently emit in the mid-infrared and can directly provide frequency comb operation at wavelengths beyond 3 μm , as discussed in the following section.

3.3 QCL-based mid-infrared DCS

QCLs are unipolar semiconductor lasers based on engineered intersubband transitions in semiconductor quantum wells, providing direct access to the mid-infrared spectral region beyond 3 μm . In QCL-based frequency combs, multiple longitudinal modes are intrinsically phase correlated through strong intracavity nonlinear interactions, primarily four-wave mixing enabled by the ultrafast gain dynamics of the active medium. This self-phase-locking mechanism allows QCLs to operate as compact, electrically pumped mid-infrared frequency comb sources without external nonlinear conversion stages. Owing to their high output power, direct long-wavelength emission, and inherently comb-like multimode operation, QCL-based OFCs are particularly well suited for DCS, especially in spectral regions that are difficult to access with DFG-based or OPO-

based approaches.

Early studies demonstrated room-temperature QCL-based frequency comb operation at wavelengths around 7 μm ⁵⁷. The device exhibited a relative bandwidth of 4.4%, indicating that four-wave mixing within the QCL cavity can spontaneously generate a frequency comb without active mode locking. Building on these advances, QCL-based DCS was subsequently demonstrated⁵⁸, with overlapping spectral bandwidths approximately 15 cm^{-1} and comb tooth spacing precision down to the millihertz level. To further improve integration and long-term stability, the team developed a chip-on architecture⁵⁹. This system operated around a wavelength of 7.5 μm , providing a 32 cm^{-1} overlapping spectral bandwidth and demonstrated broadband mid-infrared performance with a GHz-level repetition rate.

Recent progress in QCL-based DCS includes stabilization of the comb degrees of freedom. Coherent injection locking provided an all-electrical route to stabilizing QCL combs by locking the repetition frequency to a radio-frequency reference⁶⁰, while preserving comb coherence in a compact platform. More recently, mid-infrared QCL dual-comb systems have been further stabilized and frequency referenced using a single-mode QCL reference, which improves the accuracy of the frequency axis⁶¹. Optical injection has also been used to synchronize two QCL combs by locking one comb line from each source to a common master laser, thereby synchronizing their offsets and, together with electrical locking of the repetition frequencies, enabling stabilized dual-comb operation⁶².

Substantial efforts have also been devoted to extending the output power and spectral coverage of QCL-based DCS. High-power QCL frequency combs with watt-level output and center wavelengths near 8 μm have been demonstrated through dispersion-engineered active regions, achieving optical bandwidths approaching 85 cm^{-1} . When implemented in DCS configurations, these sources have enabled the generation of 215 comb lines spanning more than 70 cm^{-1} of optical bandwidth⁶³. Concurrently, QCL-based DCS was extended to different central wavelengths^{64,65}. For

high-precision spectral measurements, gapless spectral coverage has been achieved using free-running QCL-based DCS with current-tuned frequency combs. In this approach, each comb provided an optical bandwidth of 55 cm^{-1} with several hundred milliwatts of power, enabling continuous spectral coverage with a resolution of 0.001 cm^{-1} (see Fig. 7)⁶⁶.

Complementary techniques based on step-scan current tuning have also been developed to construct MHz-level frequency-axis calibration, with demonstrated operation around 1308 cm^{-1} over a 50 cm^{-1} span and GHz-level repetition rate⁶⁷. These experiments achieved MHz-level precision without requiring external phase-locking, highlighting the inherent frequency stability and electronic tunability of the QCL comb. To further extend the detection range, a long-wave infrared DCS was presented, based on QCLs emitting at a central wavelength of $9\text{ }\mu\text{m}$ with watt-level output power⁶⁸.

Taken together, QCL-based DCS provides direct access to

the molecular fingerprint region with compact, electrically pumped sources, typically operating from 5 to $10\text{ }\mu\text{m}$. Reported systems generally offer optical spans of several tens of inverse centimetres, native comb spacings of 7.5 – 15 GHz , and output powers from milliwatts to watts. Their high repetition frequencies enable rapid spectral refresh and sub-microsecond time resolution, while current tuning and spectral interleaving can improve the effective sampling interval to the megahertz or sub-megahertz level. However, the large native mode spacing limits direct sampling of narrow molecular features, and spectral interleaving improves resolution at the cost of acquisition speed and complexity. In addition, the instantaneous spectral coverage is typically narrower than that of broadband DFG- or OPO-based systems, while frequency accuracy and coherent averaging remain sensitive to phase noise, thermal and current fluctuations, and dispersion. Consequently, QCL-based DCS is particularly attractive for compact, high-power, and

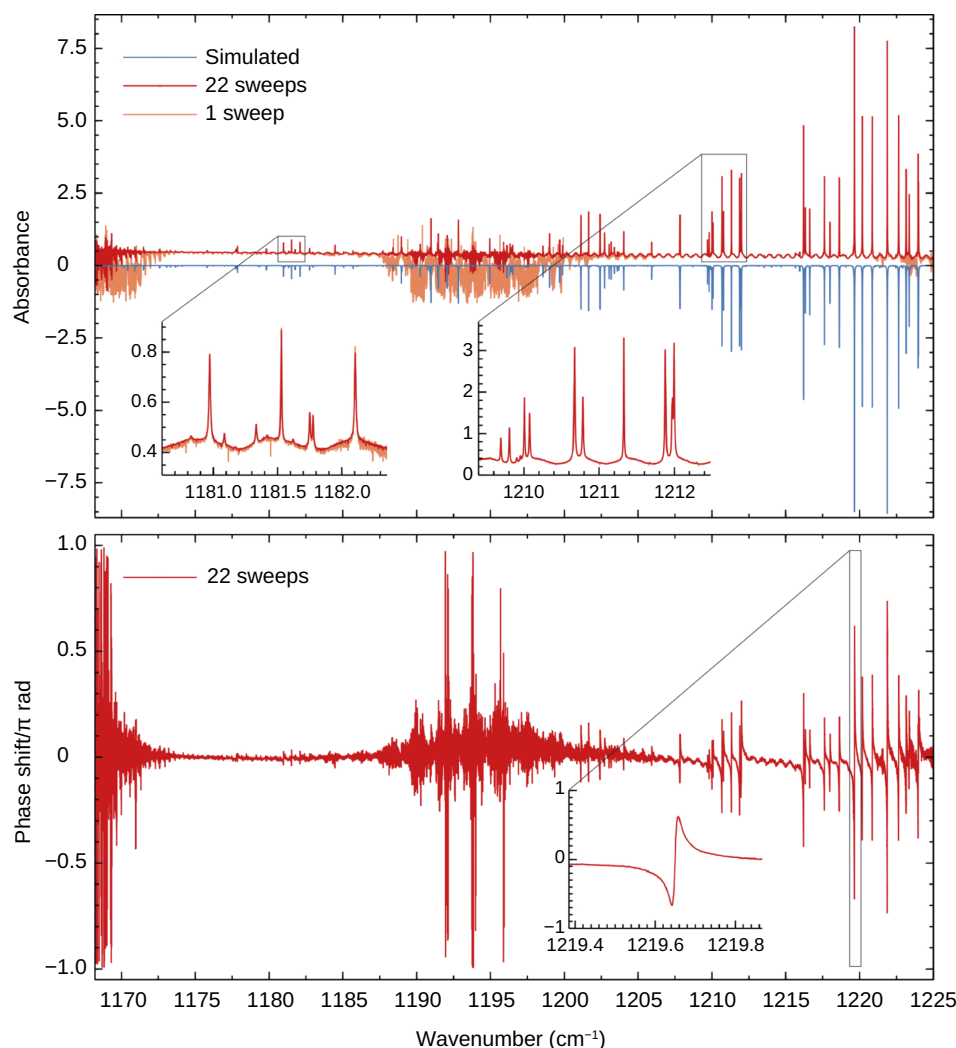


Fig. 7 | Absorption (top) and dispersion (bottom) spectra of methane. The insets show zooms of the data in the highlighted regions. Figure reproduced with permission from ref.⁶⁶, under the terms of the OSA Open Access Publishing Agreement.

high-speed sensing in selected fingerprint bands, whereas broadband and high-accuracy spectroscopy generally requires additional stabilization, calibration, or computational correction. The potential applications of QCL-based DCS have been extensively discussed in recent reviews⁶⁹.

3.4 On-chip mid-infrared DCS

Mid-infrared DCS generation on integrated photonic platforms has advanced through three representative nonlinear mechanisms: microresonator-based Kerr combs, nanophotonic supercontinuum generation (SCG), and waveguide-based DFG⁷⁰. Microresonator Kerr combs rely on cascaded four-wave mixing under continuous-wave pumping to generate dissipative Kerr solitons, which can be further broadened through careful dispersion and cavity design. SCG exploits soliton dynamics and dispersive-wave emission in dispersion-engineered nanophotonic waveguides to coherently extend near-infrared combs toward longer wavelengths. In contrast, waveguide-based DFG schemes exploit intrapulse interactions to enable broadband, phase-matched

frequency down-conversion into the mid-infrared region. Together, these approaches provide complementary routes toward chip-scale, broadband, and mutually coherent mid-infrared DCS.

Silicon-chip-based mid-infrared DCS has been realized using Kerr nonlinearity in CW-pumped microresonators (Fig. 8), generating mutually coherent combs spanning 2.6–4.1 μm ⁷¹. Due to the ultra-short cavity length, such Kerr-comb sources feature large mode spacing and high pulse energy, resulting in high SNR and fast speed. Beyond Kerr-comb implementations, nanophotonic SCG offers an alternative route toward integrated mid-infrared DCS, as illustrated in Fig. 9. This scheme employed dispersion-engineered Si_3N_4 waveguides to coherently extend near-infrared combs to 2.5–3.7 μm via soliton dynamics and dispersive-wave emission, realizing a fully integrated mid-infrared DCS⁷². To further enhance coherence and integration, a counter-propagating soliton microresonator DCS has been developed based on interleaved DFG⁷³. This scheme generated two mutually coherent mid-infrared combs centered near 3.3 μm with line spacings of 1.4–2.8 GHz. In

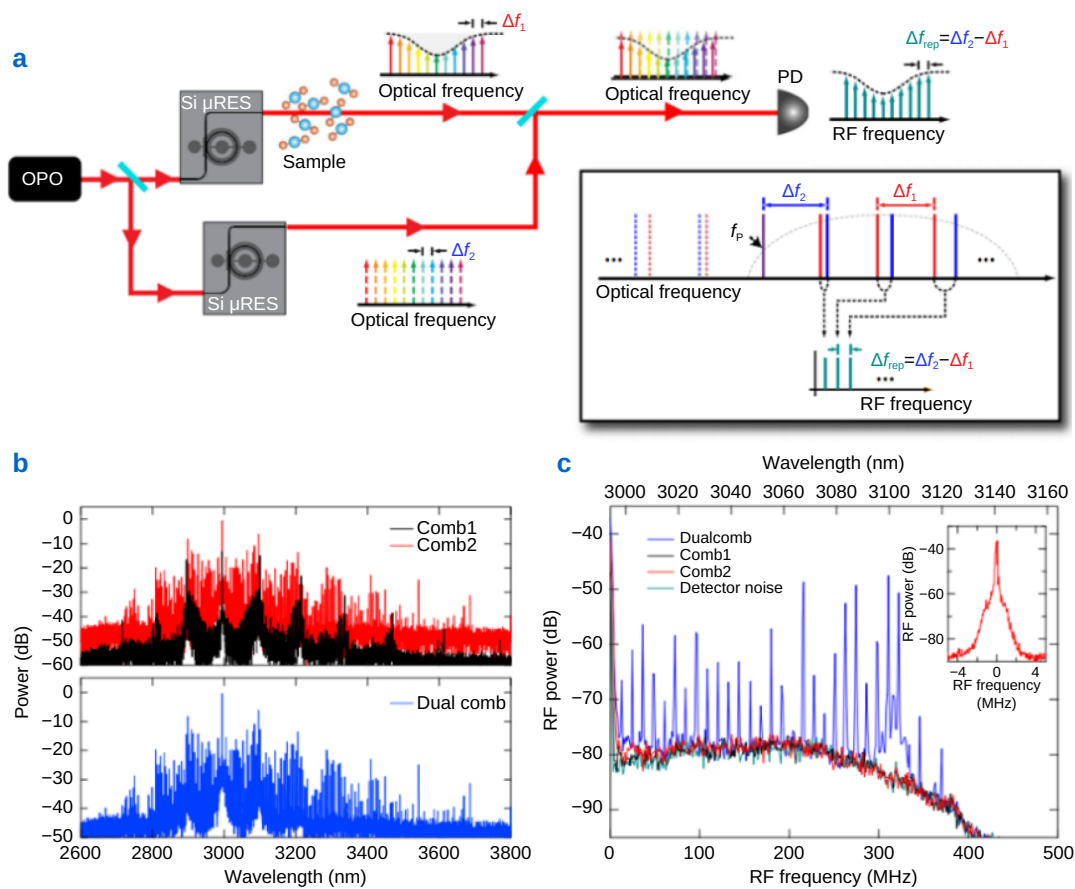


Fig. 8 | Silicon-chip-based mid-infrared dual-comb generation. (a) Experimental setup. A continuous-wave OPO pumps two independent silicon microresonators to generate mutually coherent mode-locked Kerr combs. (b) Optical spectra of the two combs (red and black) and their combined multi-heterodyne spectrum (blue). (c) RF spectrum. Figure reproduced with permission from ref.⁷¹, under a Creative Commons Attribution License.

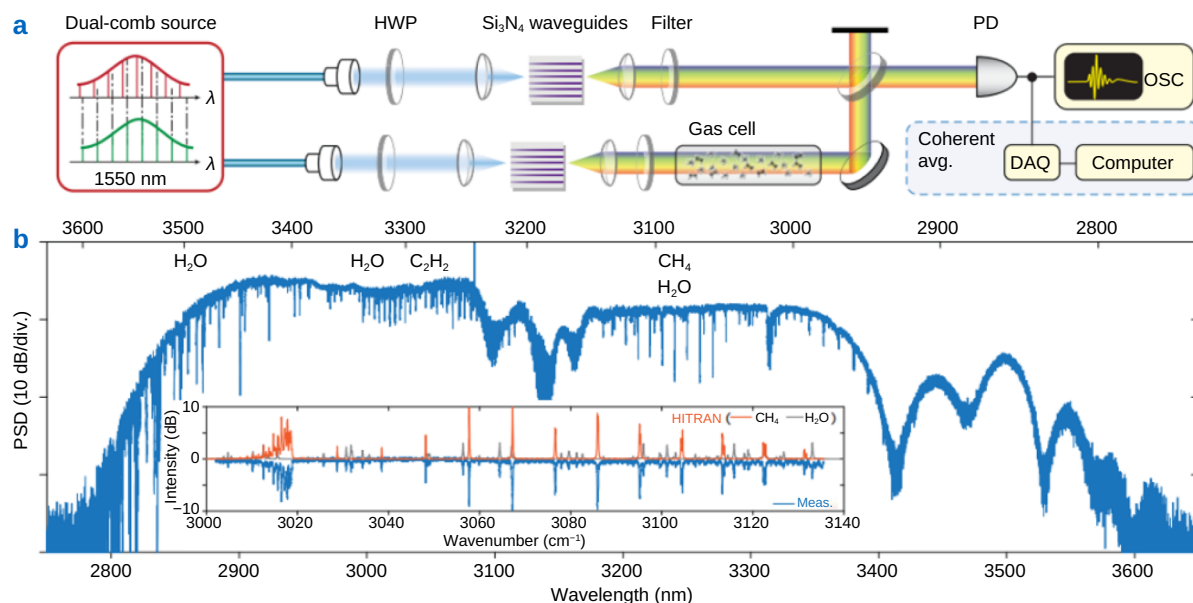


Fig. 9 | Nanophotonic supercontinuum-based mid-infrared DCS. (a) Experimental setup. (b) Retrieved mid-infrared frequency comb featuring absorption of gas species CH_4 , C_2H_2 , as well as H_2O in the circumstance. Figure reproduced with permission from ref.⁷², under the terms of the OSA Open Access Publishing Agreement.

parallel, efforts have also focused on increasing output power and spectral coverage, a waveguide-type PPLN DFG device generated mW-level mid-infrared DCS spanning 2.7–4.6 μm , which were successfully applied to gas-phase absorption spectroscopy⁷⁴. Further reduction of system complexity was achieved through IP-DFG in LiNbO_3 waveguides, yielding a compact mid-infrared DCS covering 3–4 μm (~25 THz bandwidth) with 100 MHz resolution and excellent long-term stability for methane detection⁷⁵.

Recent advances in soliton microcomb engineering have further improved the prospects of microresonator-based DCS. Although most demonstrations have been performed in the near-infrared and telecom bands, the underlying techniques are relevant to future mid-infrared and THz DCS because they improve comb stability, spectral quality, and dispersion control. For example, Fu et al. demonstrated soliton microcomb generation using cavity polygon modes in a lithium-niobate microresonator, showing the benefits of mode engineering for dispersion management⁷⁶. Liu et al. reported soliton microcombs with nearly ideal spectral envelopes in silicon oxynitride microresonators, which are promising for low-noise spectroscopy and ranging applications⁷⁷. These advances highlight the importance of soliton and dispersion control for developing compact and coherent on-chip DCS sources.

Table 4 compares integrated mid-infrared DCS platforms and their trade-offs. Kerr combs enable high per-line power and fast acquisition but typically have large comb spacing. Nanophotonic SCG provides broad and coherent spectral coverage with relatively dense sampling, while requiring

amplified femtosecond pumping and careful dispersion engineering. Waveguide-based DFG, including microcomb-assisted interleaved DFG and intrapulse DFG, offers compact and efficient wavelength conversion with flexible comb spacing, although its performance is limited by phase matching, waveguide dispersion, loss, and detector response. These advances have expanded the accessible mid-infrared range from 2.6–4.6 μm , corresponding to more than 40 THz of spectral coverage. The technological evolution from bulk-crystal DFG to integrated waveguides, nanophotonic SCG, and Kerr microresonator combs demonstrates a clear trend toward on-chip, broadband, and highly coherent mid-infrared DCS.

Taken together, a key requirement for implementing DCS is the availability of coherent frequency combs operating within the target spectral range. While OFC technology in the near-infrared band has become increasingly mature, comb generation in the mid- and far-infrared regions is still under active development. As summarized in Fig. 10, DCS demonstrations across different spectral regions exhibit pronounced variations in spectral resolution, frequency accuracy, SNR, and system-level error control, reflecting the strong dependence of DCS performance on the maturity of OFC technology in the corresponding wavelength range.

4 THz spectroscopy

THz waves, spanning approximately 0.1–10 THz, occupy the spectral region between microwaves and infrared light. In recent years, advances in optical frequency comb

Table 4 | Summary of representative On-chip mid-infrared DCS architectures.

Architecture	Spectral coverage	Resolution/comb spacing	FOM	Output power	Main strengths	Main limitations
Microresonator-based Kerr combs ⁷¹	2.6–4.1 μm	Typically tens GHz to 127 GHz	$>10^6$ Hz ^{1/2}	A few mW	Ultrafast acquisition	Large comb spacing limits high-resolution gas spectroscopy
SCG ⁷²	2.5–3.7 μm	Typically on the order of 100 MHz	$\sim 10^6$ Hz ^{1/2}	A few mW	Broad, flat, coherent coverage with dense sampling.	Requires amplified femtosecond pumping and precise dispersion control
Waveguide-based DFG ^{73–75}	2.7–4.6 μm	Typically from ~ 100 MHz to several GHz	$\sim 10^6$ Hz ^{1/2} ⁷⁵	A few mW	Compact and efficient MIR wavelength conversion; IP-DFG can be offset-free	Waveguide dispersion and detector response limit bandwidth and SNR

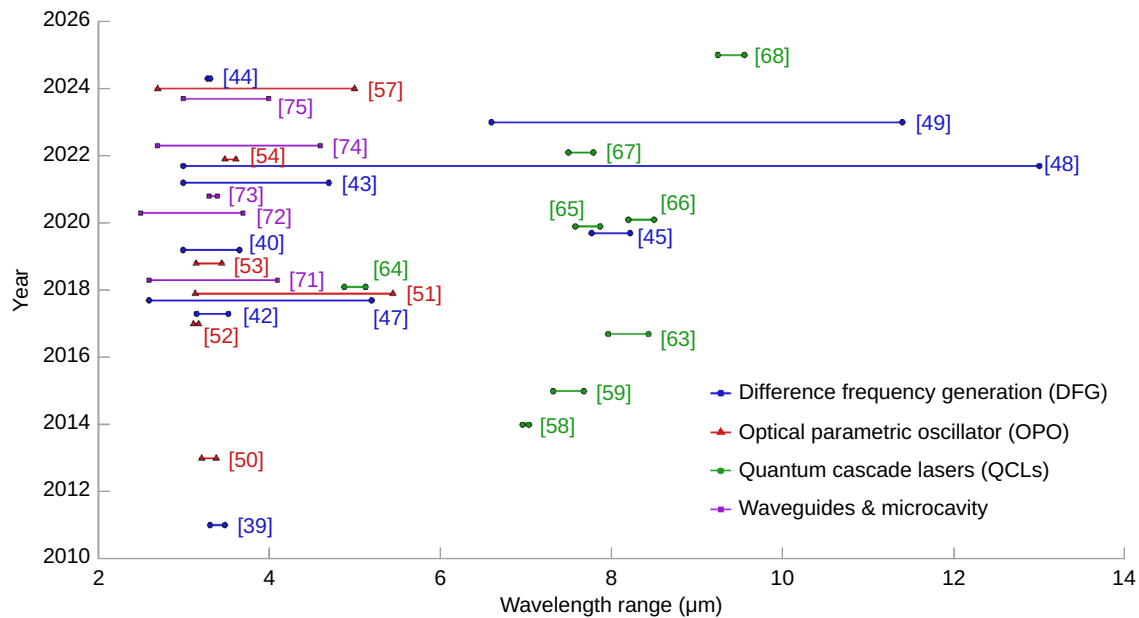


Fig. 10 | Spectral coverage of DCS demonstrations. Different colors denote different source platforms, including DFG-based mid-infrared DCS^{39,40,42–49}, OPO-based mid-infrared DCS^{50–55}, QCL-based mid-infrared DCS^{58–60,63–68}, and waveguide- or microcavity-based mid-infrared DCS^{71–75}.

technology have opened a transformative route into the THz domain. Broadband THz frequency combs are currently realized via two principal technological approaches, each offering distinct advantages. The first relies on optical down-conversion: femtosecond lasers drive photoconductive antennas (PCAs) or EO crystals to generate THz combs with wide spectral coverage (e.g., 0.1–4 THz) and dense line spacing, well suited for broadband, high-resolution spectroscopy. The second approach is based on direct THz generation, most prominently using fully stabilized QCL combs. These sources provide high power per comb line and exceptional fractional frequency stability, reaching the 10^{-12} level, making them particularly advantageous for high signal-to-noise measurements, especially in strongly absorb-

ing molecular bands. THz frequency combs generated by both methods enable direct frequency comparison between THz clocks and optical clocks, effectively linking THz frequencies to the optical standards traceable to the International System of Units (SI).

4.1 Broadband THz frequency comb spectroscopy

Broadband THz frequency combs are predominantly generated via optical-to-THz conversion, in which femtosecond optical frequency combs serve as the pump sources. As a result, the temporal and spectral properties of the THz comb are fundamentally inherited from those of the driving optical comb. In the time domain, the pulse duration of the

femtosecond comb determines the achievable THz spectral bandwidth: shorter optical pulses correspond to broader THz spectra. Meanwhile, precise control of the pulse repetition rate and timing sequence is essential for constructing stable THz frequency combs, and it underpins high-precision sampling techniques such as asynchronous optical sampling (ASOPS). In the frequency domain, the stability of the optical comb directly governs the linewidth, phase coherence, and long-term stability of the generated THz comb. Through mode-locking and frequency-stabilization techniques², an ultrastable and highly coherent optical frequency comb can be realized. These attributes are subsequently transferred to the THz regime via nonlinear frequency conversion, yielding a THz frequency comb with exceptionally narrow linewidth and well-defined absolute frequencies. This direct transfer of coherence and frequency accuracy establishes a solid foundation for ultrahigh-precision THz spectroscopy, precision frequency metrology, and coherent detection applications.

THz dual-comb spectroscopy (THz-DCS) was developed to overcome the resolution limitations inherent to conventional mechanical delay-line systems. By employing two frequency combs with slightly detuned repetition rates to implement ASOPS, THz-DCS eliminates mechanical scanning and maps the fast THz time-domain signal into a radio-frequency domain. This strategy significantly enhances both spectral resolution and acquisition speed. The performance of THz-DCS critically relies on precise stabilization of the optical combs and effective suppression of their relative timing jitter. To maintain long-term mutual coherence, advanced techniques such as adaptive sampling and common-path single-cavity dual-comb (SCDC) laser architectures are employed. These approaches ensure stable multi-heterodyne detection and enable high-precision, Doppler-limited spectroscopic measurements across the THz band. A pioneering demonstration was reported in 2006 by Yasui et al.⁷⁸, who realized broadband multimode THz frequency comb detection using a multi-heterodyne scheme. They achieved a comb spacing of 81.8 MHz and a frequency accuracy of 2.5×10^{-7} , marking the first successful extension of near-infrared dual-comb spectroscopy into the THz regime.

In 2015, Finneran et al.⁷⁹ reported a THz-DCS system that simultaneously achieved decade-spanning bandwidth and kilohertz-level comb linewidth, effectively addressing the longstanding trade-off between spectral coverage and frequency accuracy in the THz regime. Their comb source covered 0.15–2.4 THz with an 80 MHz mode spacing and exhibited a single-comb-tooth linewidth of 3.7 kHz, corresponding to a relative precision of 1.8×10^{-9} . In measurements of water vapor transitions, the system attained an accuracy of 6.1×10^{-8} , reaching Doppler-limited precision (Fig. 11(a, b)). Notably, this performance was realized in a fully room-temperature platform, establishing a practical and powerful tool for frontier applications such as astro-

physics and high-resolution molecular spectroscopy.

To overcome the limitation imposed by the discrete comb mode spacing in broadband spectral sampling, Yasui et al.⁸⁰ introduced a swept THz-DCS interleaving technique in 2017. By finely tuning the comb parameters and interleaving successive spectra, they achieved an effective sampling interval of 2.5 MHz—equal to the comb-tooth linewidth—and a frequency accuracy of 8.39×10^{-7} in measuring the $1_{10} \leftarrow 1_{01}$ rotational transition of water vapor (10 Pa) at 0.557 THz (Fig. 11(c–e)). This approach significantly enhanced spectral point density without sacrificing accuracy. Building on these advances, Konnov et al.⁸¹ generated an ultrabroadband THz frequency comb spanning 1.5–45 THz while maintaining an 80 MHz comb spacing and a 10 MHz effective resolution through spectral interleaving. In measurements of CO₂ and C₂H₂ gases at 4 mbar, the system successfully resolved absorption features with linewidths as narrow as 66 MHz (Fig. 11(f)), demonstrating strong potential for rapid, high-precision molecular fingerprinting across an exceptionally wide spectral range.

Technological advances such as SCDC lasers and adaptive compensation schemes—originally developed for optical dual-comb systems—have now been successfully translated to the THz domain. Their implementation has markedly reduced the complexity of THz frequency comb systems by eliminating the need for fully independent comb stabilization. The intrinsic “passive coherence” of SCDC architectures provides a robust platform for high-precision, high-resolution THz measurements. For example, Hu et al.⁸² realized broadband THz spectroscopy spanning 0–2 THz using an SCDC-based system. By incorporating an intracavity Lyot filter to tailor the gain spectrum and compensate dispersion, they achieved stable dual-comb operation and high-precision measurements of rotational transitions in acetonitrile (CH₃CN). For 39 resolved absorption peaks, the measured frequencies deviated by only 563 MHz from the JPL database values, demonstrating both practical system simplification and reliable broadband detection capability. Further improvements in long-term stability were demonstrated by Nakajima et al.⁸³, who implemented an SCDC configuration based on a mechanically shared, fully polarization-maintaining fiber laser structure. This design achieved an Allan deviation as low as 0.02 Hz at a 1 s averaging time and enabled high-precision identification of water vapor absorption lines across the 0.1–1.5 THz range. These developments highlight the effectiveness of SCDC-based architectures in enhancing coherence, stability, and practicality for THz dual-comb spectroscopy.

To mitigate the non-uniform sampling arising from frequency-offset drift in SCDC systems, Chen et al.²⁶ proposed an adaptive sampling scheme based on a free-running SCDC fiber laser. As shown in Fig. 12(a, b), their method dynamically corrected timing fluctuations without requiring tight phase locking, enabling mode-resolved THz dual-comb spectroscopy. The system achieved a spectral

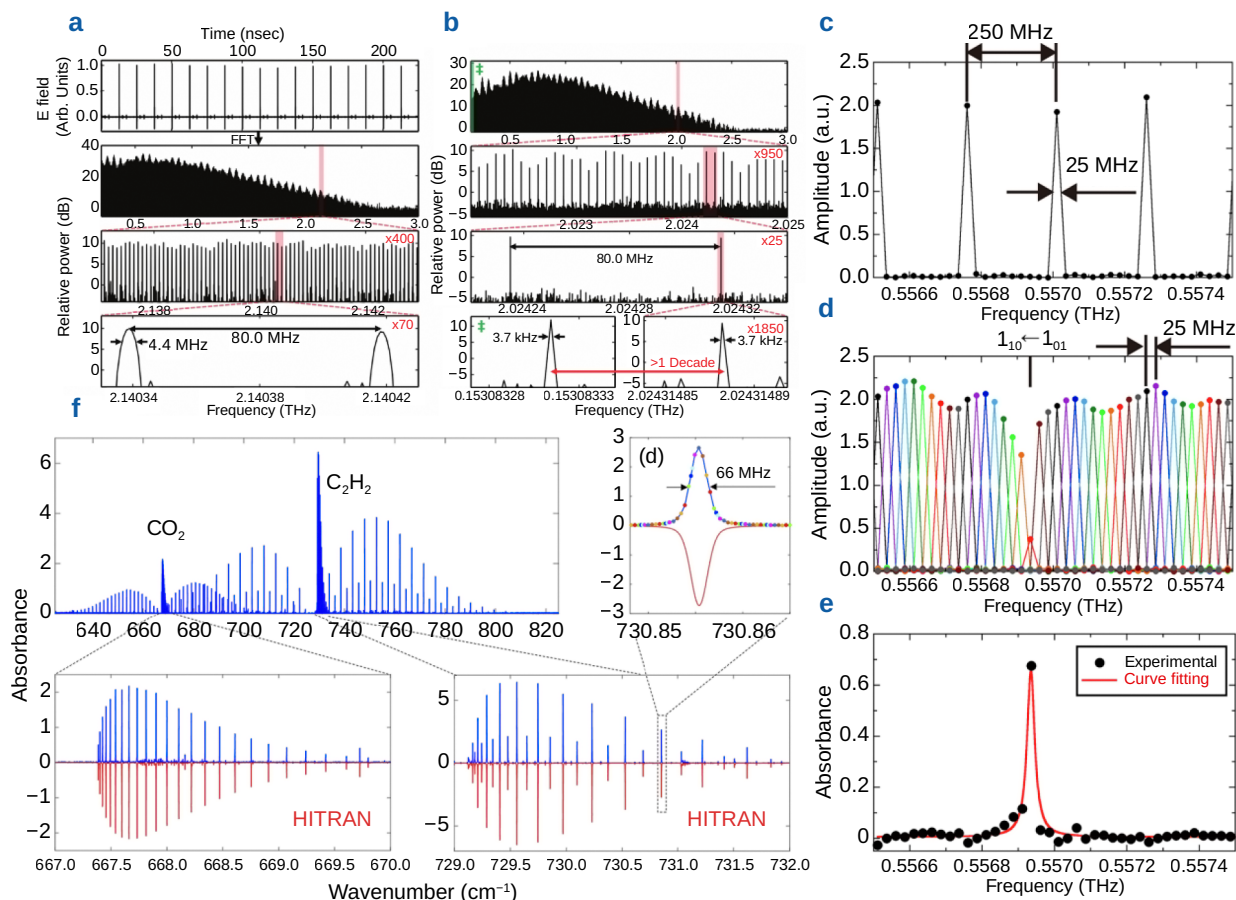


Fig. 11 | Schematic diagram and typical results of a dual-comb system based on mode-locked lasers. (a) Fourier transform of a 225 ns THz pulse train in delay time yields a frequency comb that extends up to 2.4 THz, with a 4.4 MHz tooth width. (b) The tooth width of the THz comb can be reduced to 3.7 kHz by extending the time-domain measurement to 268 μ s in delay time. (c) The THz comb spectrum near 0.557 THz. (d) Amplitude spectrum of a spectrally interleaved THz comb over the same frequency range. By sweeping the repetition rate and superimposing 10 measurements, the discrete spectrum is effectively filled in, revealing a clear absorption dip. (e) Pressure broadening characteristics of the rotational transition $1_{10} \leftarrow 1_{01}$ in water vapour. (f) High-precision mid-infrared application: By combining eight sets of measurements from frequency-offset combs (different colored dots), the effective spectral sampling interval is densified from 80 MHz to 10 MHz, precisely resolving the absorption profile of acetylene (C_2H_2) at 730.85 cm^{-1} with a linewidth of 66 MHz. Figure reproduced with permission from: (a, b) ref.⁷⁹, American Physical Society; (c–e) ref.⁸⁰, (f) ref.⁸¹, under a Creative Commons Attribution License.

resolution of 48.8 MHz and a comb-tooth linewidth of 4.88 MHz, demonstrating that high-quality THz-DCS can be realized even with a simplified stabilization strategy. In contrast, Xia et al.⁸⁴ pursued enhanced long-term stability by incorporating real-time clock correction into a THz-DCS platform. Their approach extracted optical beat-frequency signals and applied loose active frequency stabilization to compensate for slow drifts, substantially improving operational robustness. The system maintained continuous operation for over 1200 hours and achieved a spectral resolution of 66.14 MHz, an ultranarrow comb-tooth linewidth of 1.9 kHz, and a relative frequency accuracy of 1.7×10^{-9} (Fig. 12(c)). Together, these strategies—adaptive sampling and real-time clock correction—illustrate complementary pathways toward simplifying THz-DCS architectures while preserving high resolution, narrow linewidth, and long-term

stability.

4.2 Electro-optic comb-based THz DCS

According to existing reports, THz frequency combs generated from mode-locked lasers (MLLs) typically achieve a relative frequency precision on the order of 10^{-9} , corresponding to comb linewidths in the kilohertz range. Achieving higher precision requires tightly stabilizing the femtosecond optical comb to an ultra-stable CW reference laser. While this strategy can substantially improve long-term frequency accuracy, it introduces significant technical challenges. In particular, the broadband phase noise intrinsic to femtosecond combs is difficult to fully suppress due to the finite bandwidth of electronic phase-locked loops. As a result, residual noise limits coherence transfer, and the overall

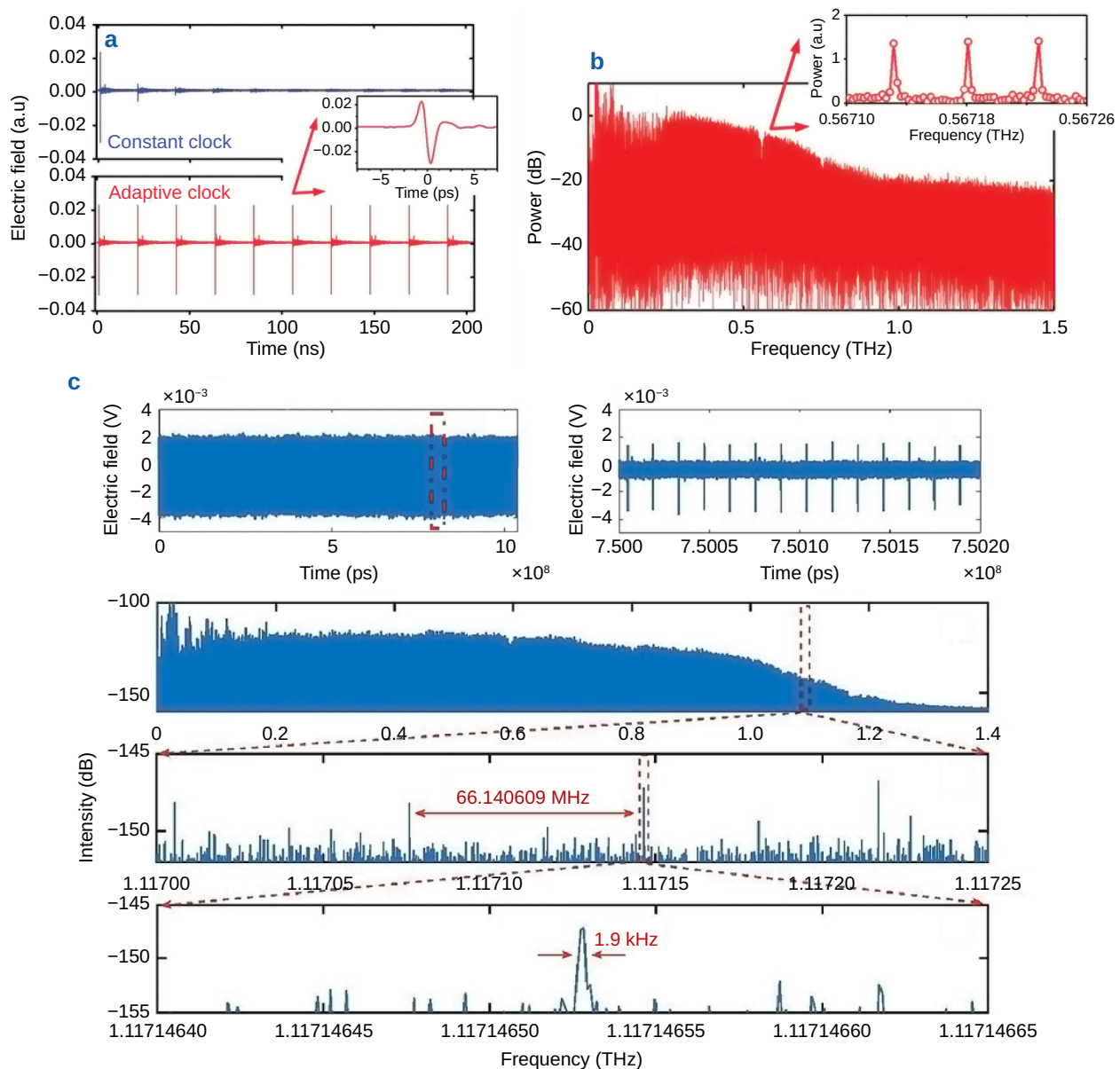


Fig. 12 | Adaptive THz comb spectrum. (a) Comparison of time-domain waveforms averaged 100000 times under different sampling clocks. (b) Comb-mode-resolved THz spectrum obtained in ambient air at room temperature and pressure. (c) Fourier transform of a frequency comb synthesized from 68524 consecutive THz pulses using adaptive clocking, covering up to 1.2 THz with a comb linewidth of 1.9 kHz and a peak dynamic range of 35 dB. Figure reproduced with permission from: (a, b) ref.²⁶, (c) ref.⁸⁴, under a Creative Commons Attribution license.

system becomes considerably more complex, bulky, and less practical for widespread deployment. In addition, the cavity-defined repetition rate of MLLs inherently restricts the tunability of the comb line spacing, typically to within $\sim 1\%$ of the nominal repetition frequency. This limited flexibility constrains their applicability in scenarios that demand finely adjustable frequency grids, such as resolving closely spaced hyperfine transitions in gas-phase molecules.

Recently, EO combs have emerged as a compelling alternative. Because they are directly synthesized from a single-frequency CW laser—often referenced to an atomic

clock—their frequency stability is passively inherited from the microwave or optical reference. Furthermore, EO combs offer wide and continuous tunability of both comb spacing and carrier frequency, together with a comparatively simple and robust architecture. These features make EO-comb-based THz systems particularly attractive for high-precision, flexible, and field-deployable spectroscopic applications. As an early demonstration of the potential of EO combs in the THz domain, Jerez et al.⁸⁵ developed an absolute-frequency THz dual-comb spectrometer based solely on electro-optic modulators and standard telecom components. The

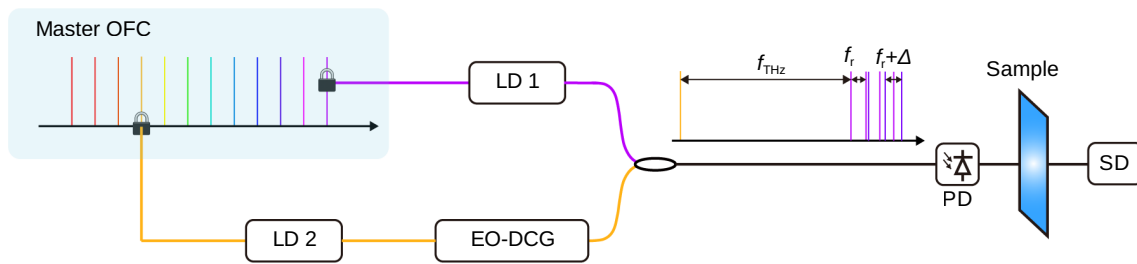


Fig. 13 | Experimental setup and sketch of the generation process of the electronically synthesized THz dual-comb. A master OFC serves as a frequency ruler for coherent mode selection via OIL. Phase modulating a first optical mode in two paths generates an optical dual-comb. The signal is photomixed with a second filtered mode from the master OFC to create an ultra-low phase noise THz dual-comb that probes a sample. Overlapping the THz dual-comb in a Schottky diode generates a downconverted comb in the RF domain that can be digitized and processed to obtain the imprinted spectral profile. In alphabetical order, LD, laser diode; PD, photodetector; SD, Schottky diode.

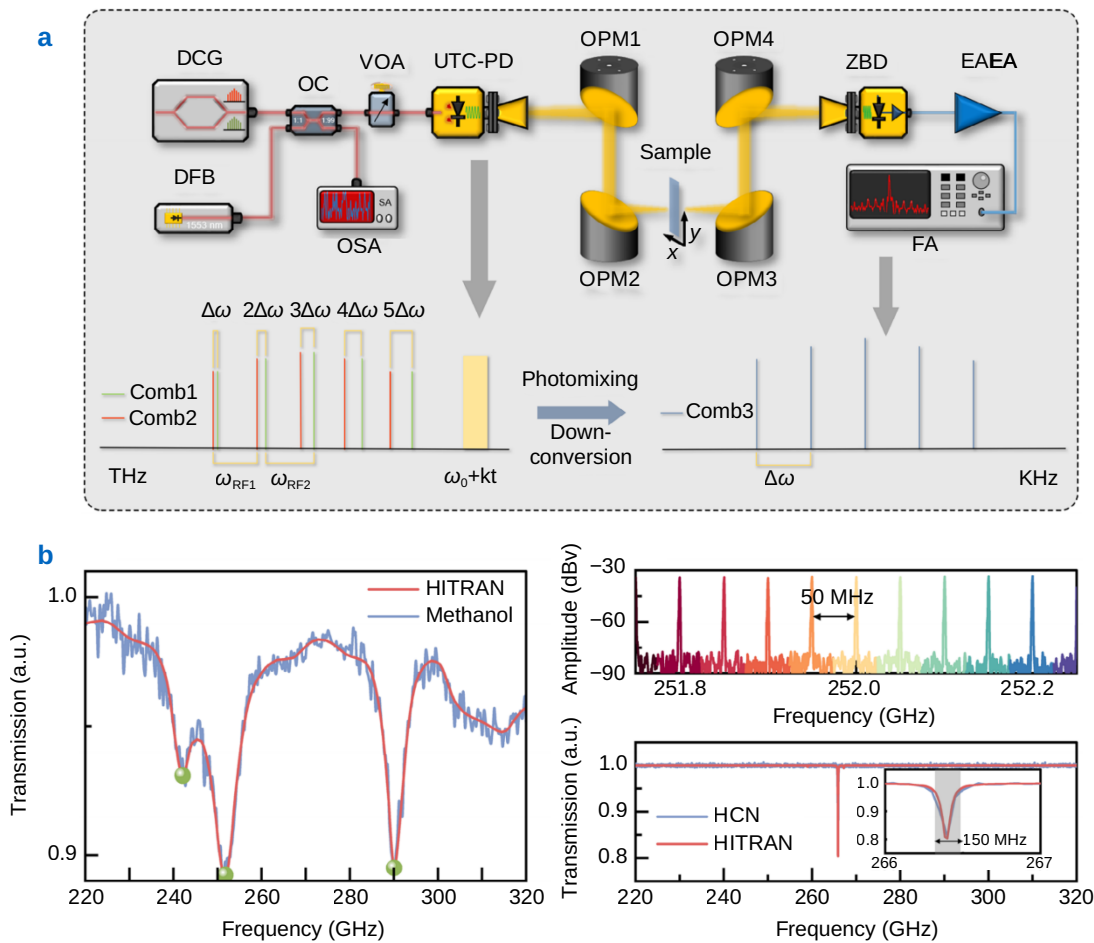


Fig. 14 | Dual electro-optic comb THz spectroscopy. (a) Schematic diagram of photonic THz spectrum with swept-frequency dual combs. (b) Transmission spectra of HCN vapor in 0.22–0.32 THz. Figure reproduced with permission from ref.86, under a Creative CommonsAttribution License.

working principle is shown in Fig. 13. In this design, a single optical line filtered from a master frequency comb via optical injection locking serves as the seed for the electro-optic generation of two new combs, designated as the probe and the local oscillator. These two combs, together with another coherent line selected from the same master comb, are photomixed in a photoconductive antenna to produce a

THz dual-comb characterized by narrow linewidth and comb teeth frequencies traceable to a radio-frequency standard. The system provides a tunable center frequency from 0.09596 to 0.49596 THz, a fixed comb spacing of 100 MHz, a temporal resolution of 10 μ s, and absolute frequency accuracy⁸⁵. Long-term stability characterization reveals a frequency drift of less than ± 2.5 Hz over 90 minutes, with

the 3 dB linewidth below the resolution bandwidth of the spectrum analyzer—indicating high mutual coherence. The overlapped Allan deviation exhibits a fractional frequency instability between white frequency modulation and flicker frequency modulation. With an integration time of 400 μ s, the system achieves an average signal-to-noise ratio of 102 over 75 spectral lines, corresponding to a figure of merit of 3.83×10^5 at 1 second. This architecture eliminates the need for dual stabilized femtosecond lasers, offering a compact, cost-effective, and highly flexible solution for high-resolution THz spectroscopy.

Building on the capabilities of EO combs for precise spectral measurements, Fang et al.⁸⁶ demonstrated a swept dual-comb hyperspectral imaging system in the THz regime by combining a static dual EO comb source with a linearly swept CW mixer. Operating over the 0.22–0.32 THz range, the system achieved a frequency resolution of 50 MHz together with millimeter-scale spatial resolution. This configuration enabled simultaneous material identification—demonstrated through the detection of methanol and hydrogen cyanide absorption features—and high-resolution imaging (Fig. 14(a, b)), highlighting the potential of dual-comb techniques for integrated spectroscopic imaging in the THz band.

In a different strategy, Stroud et al.⁸⁷ developed a dual chirped-pulse EO frequency comb system for THz spectroscopy. By employing chirped pulses with distinct bandwidths in the local oscillator and detection arms, and applying phase-sliding signal processing, they synthesized approximately 3180 effective comb lines in the radio-frequency domain. This approach enabled the simultaneous acquisition of steady-state absorption spectra and transient kinetic responses of formic acid in the 0.3–0.6 THz region, successfully resolving densely spaced and partially overlapping rotational transitions.

Despite these advances, EO-comb-based THz implementations remain limited in bandwidth and intrinsic comb line density, which are often restricted to only a few dozen physical comb modes in the THz domain. Extending the spectral coverage typically requires broadband femtosecond EO combs generated through nonlinear spectral broadening. However, such nonlinear processes tend to introduce excess phase and amplitude noise, leading to comb linewidth broadening and degraded mutual coherence. These trade-offs between bandwidth expansion and spectral purity continue to pose key challenges for high-precision, broadband THz EO-comb systems.

A strategy to overcome these limitations is to use spectral multiplication in a multi-wavelength laser (MWL), thereby avoiding the conflict between noise and bandwidth⁸⁸. In their scheme, a narrowband EO comb (with a free spectral range of ~ 1 GHz) is optically injected into a semiconductor MWL monolithically integrated with a phase-controlled feedback cavity (Fig. 15(a)). Owing to the intrinsic mode coupling within the MWL, the injected comb is spectrally

multiplied to multiple longitudinal modes separated by up to 1.3 THz (Fig. 15(b)), effectively generating several sub-combs that inherit the coherence of the original EO comb. By tuning the feedback phase on a nanosecond timescale, the system achieves agile switching between multiplied combs centered at different THz-range offsets (e.g., 26 GHz, 54 GHz, and 1.3 THz) with an extinction ratio exceeding 30 dB (Fig. 15(c, d)). Moreover, by adding an extra tone to the injected comb, cascaded phase locking among the multiplied sub-combs can be realized, yielding beat notes with sub-hertz linewidths. This approach demonstrates that THz-range comb multiplication can be achieved without nonlinear spectral broadening, thereby preserving the low phase noise of the EO comb while significantly extending its effective bandwidth (Fig. 15(e)).

Addressing the same bandwidth and noise challenges from a different perspective, a broadband THz frequency comb generated by a photoconductive antenna (PCA) driven with a low-noise near-infrared EO comb was recently reported⁸⁹ (Fig. 16(a)). The central innovation is a combined self-frequency-shifting and spectral-filtering scheme that efficiently suppresses the broadband phase noise of the EO comb, thereby eliminating the need for complex active phase-locking loops. This strategy enables the simultaneous realization of ultrawide spectral coverage (0.05–4 THz) (Fig. 16(b)), an ultranarrow comb linewidth of 0.3 Hz, and outstanding frequency stability of 8.6×10^{-14} at 1 s averaging time (Fig. 16(e)). Through asynchronous optical sampling, approximately 36000 comb lines were resolved across the THz band, demonstrating both high spectral density and long-term coherence. In addition, the system provides rapid and wide-range tunability of the comb spacing, further enhancing its flexibility for precision measurements. By combining broadband coverage, sub-hertz linewidth, and exceptional stability within a comparatively simplified architecture, this work establishes a metrology-grade platform for broadband THz frequency combs, offering substantial potential for high-resolution spectroscopy and precision frequency metrology.

4.3 THz-QCLs

Since its theoretical inception of QCLs by Kazarinov and Suris in 1971⁹⁰, it has undergone substantial development, particularly in the THz spectral region. In 1994, Faist and his collaborators at Bell Labs successfully developed the first QCL based on the InGaAs/AlInAs/InP material system⁹¹. A major milestone was reached in 2002, when Köhler et al.⁹² successfully demonstrated the first THz QCL. QCLs have since emerged as promising sources for THz generation and frequency comb operation due to the strong four-wave mixing (FWM) nonlinearity induced by spatial hole burning (SHB), along with a balance between Kerr nonlinearity and group velocity dispersion (GVD)^{93,94}. However, free-running THz QCLs face challenges as low-noise frequency

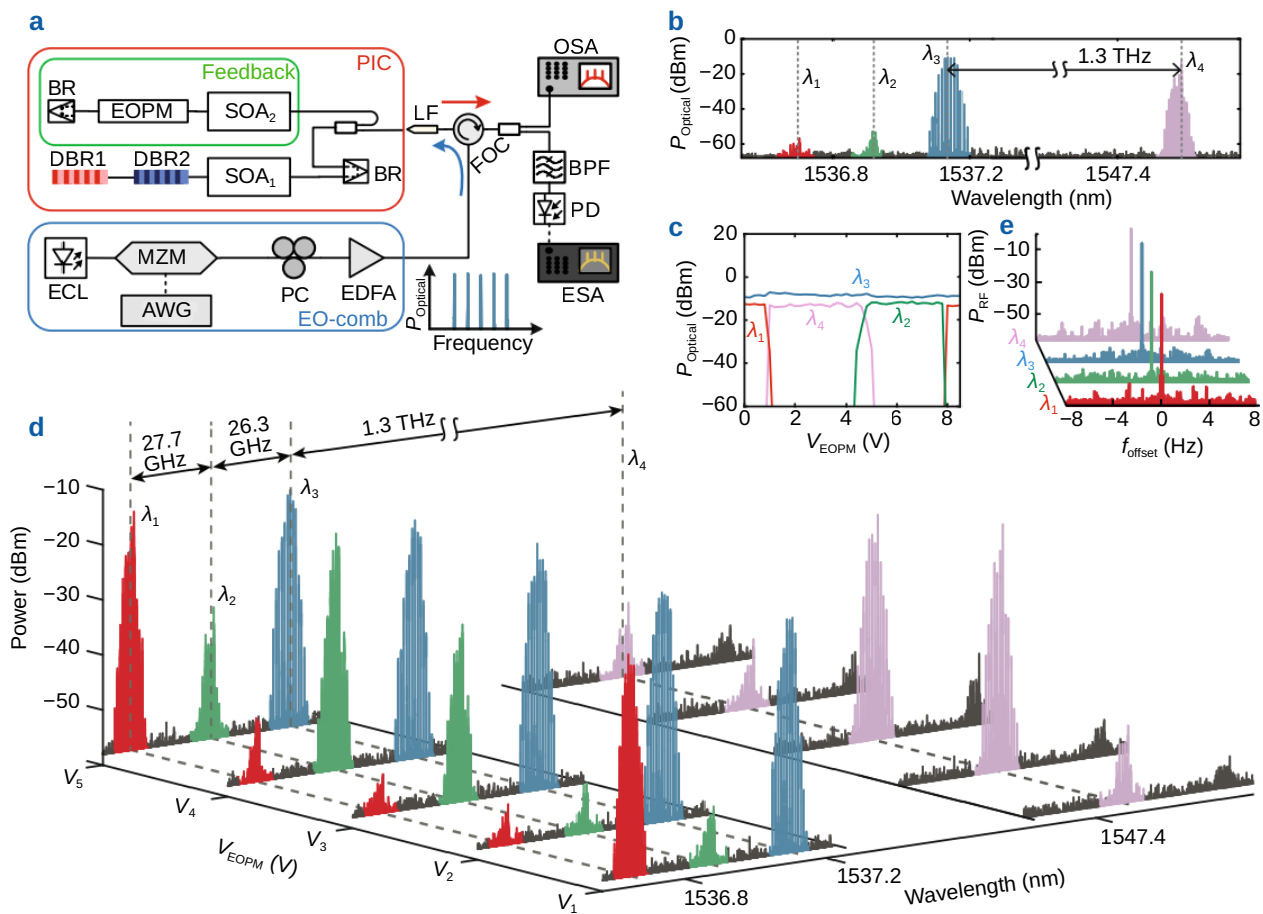


Fig. 15 | THz-range agile frequency comb spectral multiplication using a feedback-controlled semiconductor MWL. (a) Experimental setup. To generate the EO-comb, the light from an external cavity laser (ECL) is sent through a Mach-Zehnder modulator (MZM, $V_{\pi} \approx 6$ V) driven by an arbitrary waveform generator (AWG). The polarization and strength of this injected signal is adjusted with a polarization controller (PC) and an EDFA, respectively. The light is coupled in and out of the photonic integrated circuit (PIC) containing the MWL with feedback cavity via a lensed fiber (LF). A high-resolution (5 MHz RBW) optical spectrum analyzer (OSA) records the output spectrum. An optical bandpass filter (BPF) followed by a high-speed photodiode (40 GHz BW) is used to select a sub-comb which is then analyzed by an electrical spectrum analyzer (ESA). (b) Optical spectrum of the re-generated and multiplied combs when the feedback is off ($I_{SOA2} = 0$ mA) and the narrowband comb is injected around λ_3 . The dashed lines indicate the wavelengths λ_1 to λ_4 of the residual longitudinal modes of the MWL. Without feedback, the frequency comb is multiplied at around λ_4 . (c) Optical power of the various sub-combs as a function of the voltage, V_{EOPM} , applied to the EOPM for $I_{SOA2} = 21$ mA and comb injection around λ_3 . Comb switching occurs between the multiplied combs at around λ_1 , λ_2 , and λ_4 . (d) Optical spectral for various V_{EOPM} values: $V_1 = 0$ V, $V_2 = 3$ V, $V_3 = 4.6$ V, $V_4 = 7$ V, and $V_5 = 8$ V. (e) RF spectrum of the resulting beating between the tones of each comb (multiplied and re-generated). The low (< 1 Hz) linewidth, limited by the RBW of the ESA (1 Hz), shows that the coherence of the injected comb is preserved after the injection. Figure reproduced with permission from ref.⁸⁸, under a Creative Commons Attribution License.

comb sources due to phase noise originating from instabilities in both carrier frequency and repetition rate. Achieving stable comb operation requires minimizing GVD^{95,96}, with methods such as Gires–Tournois interferometers or double-chirped mirrors used to adjust the overall dispersion profile^{97,98}. Active stabilization techniques have been developed to stabilize the repetition frequency and enhance mode stability and spectral coverage^{99–103}. Phase-locking strategies further stabilize the offset frequency^{104,105}. Furthermore, passive stabilization techniques, such as integrating a saturable absorber into the laser cavity, promote stable pulse

oscillation by selectively absorbing weaker background components^{106,107}.

Unlike conventional mode-locked lasers, THz-QCL combs predominantly rely on ultrafast gain dynamics and intracavity four-wave mixing to establish phase coherence among longitudinal modes. The picosecond-scale carrier lifetime in QCL active regions enables efficient mode coupling, while the interplay between SHB-induced gain modulation, Kerr nonlinearity, and waveguide dispersion governs comb formation and stability^{94,108,109}. Consequently, even small residual GVD can disrupt phase-matching

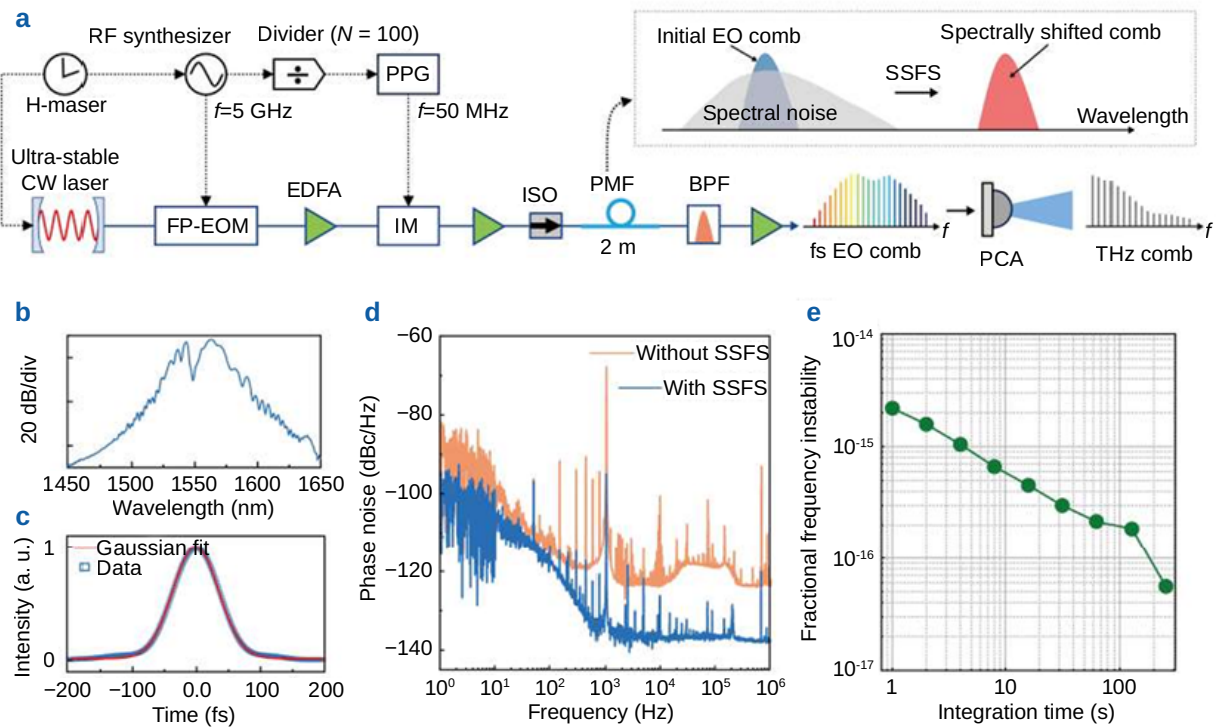


Fig. 16 | Terahertz (THz) comb generation. (a) Experimental setup. CW, continuous-wave; RF, radio-frequency; H-maser, hydrogen maser; PPG, picosecond pulse generator; FP-EOM, Fabry-Pérot electro-optical modulator; EDFA, erbium-doped fiber amplifier; IM, intensity modulator; ISO, fiber-coupled isolator; PMF, polarization-maintaining fiber; BPF, band-pass filter; PCA, photoconductive antenna; SSFS, soliton self-frequency shift; f , frequency; N , number of divisors. (b, c) Spectrum and autocorrelation trace of the femtosecond (fs) EO comb. The autocorrelation data (blue) are fitted by a Gaussian curve (red). (d) Phase noise spectra of the EO comb, measured with and without SSFS and spectral filtering. (e) Allan deviation of the EO comb. The cartoon in the dashed box illustrates the spectral separation (using SSFS) between the initial EO comb and the amplified spontaneous noise, achieved by SSFS in a 2-m PMF. Figure reproduced with permission from ref.⁸⁹, under a Creative Commons Attribution License.

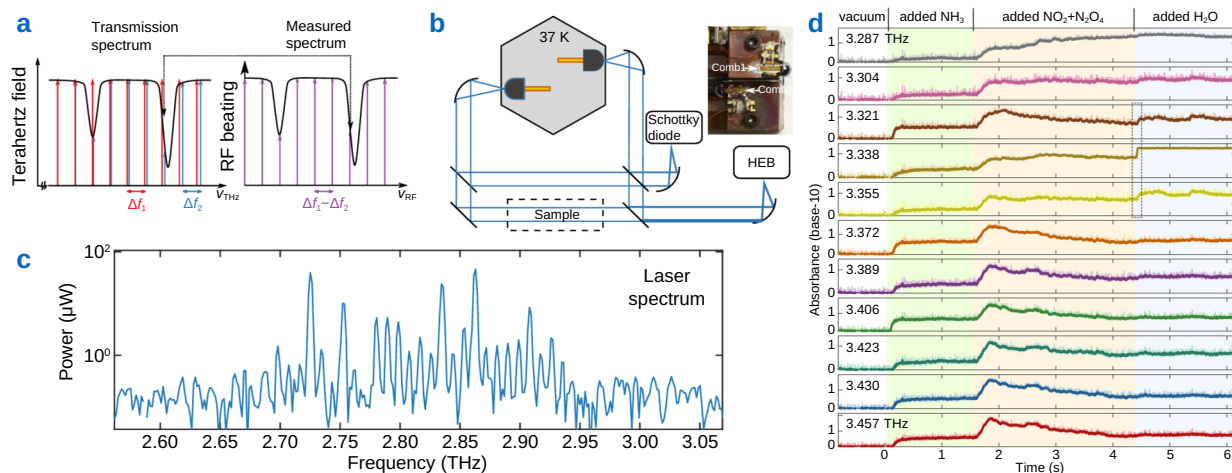


Fig. 17 | Broadband THz QCL DCS. (a) Multiheterodyne spectroscopy using two frequency combs with slightly different repetition rates. (b) Experimental setup. Inset shows real laser frequency combs on the copper mount, both of which are silicon lens-coupled. (c) Lasing spectrum of one device under comb operation. (d) Time-resolved decadic (base-10) absorbance retrieved from the dual-comb spectrogram at the four phases of the experiment. Bold lines are 10-point moving averages. Figure reproduced with permission from: (a–c) ref.¹¹⁰, under the terms of the OSA Open Access Publishing Agreement; (d) ref.¹¹¹, American Chemical Society.

conditions, resulting in comb state degradation or transition into high-phase-noise regimes. This sensitivity becomes increasingly pronounced as the spectral bandwidth is extended toward octave-spanning operation.

Although THz QCLs are compact semiconductor sources of high-power THz radiation, their narrowband emission makes them unsuitable for complex spectroscopy. To enhance the bandwidth of THz QCLs, Qing Hu et al. applied visible wavelength dispersive compensation technology to THz QCLs, demonstrating that arbitrary gain media can be fabricated to generate passive comb structures. Such combs cover a frequency range of nearly 500 GHz, featuring over 70 lines with a frequency of 3.4 THz, achieving performance improved by nearly an order of magnitude compared to active mode locking methods¹¹⁰. In Fig. 17(a–c), for THz QCL DCS, the team demonstrated the first multi-heterodyne spectroscopy using two THz QCL combs. Within a 250 GHz spectral range, they achieved an average signal-to-noise ratio (SNR) of 34 dB when using cryogenic detectors and 24

dB with room-temperature detectors, completing the measurement in just 100 μs ¹¹⁰. Further, the team expanded the potential of this technology in the THz region for gas spectroscopy by achieving, for the first time, millisecond-level time resolution measurements of gaseous molecular samples using QCL-based THz-DCS, thereby demonstrating the feasibility of utilizing QCL OFC for high-time-resolution THz-DCS¹¹¹, as shown in Fig. 17(d).

To overcome the limitation of THz QCL lacking a sensitive and fast detector, Jerome Faist et al. developed a dual comb structure that does not require a fast detector, thereby reducing the footprint of the device and eliminating additional optical components. In Fig. 18(a–c), the team utilized one of the combs as a detector to measure the multi-heterodyne beatnote signals from two sets of freely running THz QCL frequency combs, fully exploiting the unique characteristics of the quantum cascade active region. Up to 30 modes can be detected, corresponding to a spectral bandwidth of 630 GHz for the dual-comb configuration¹¹². While

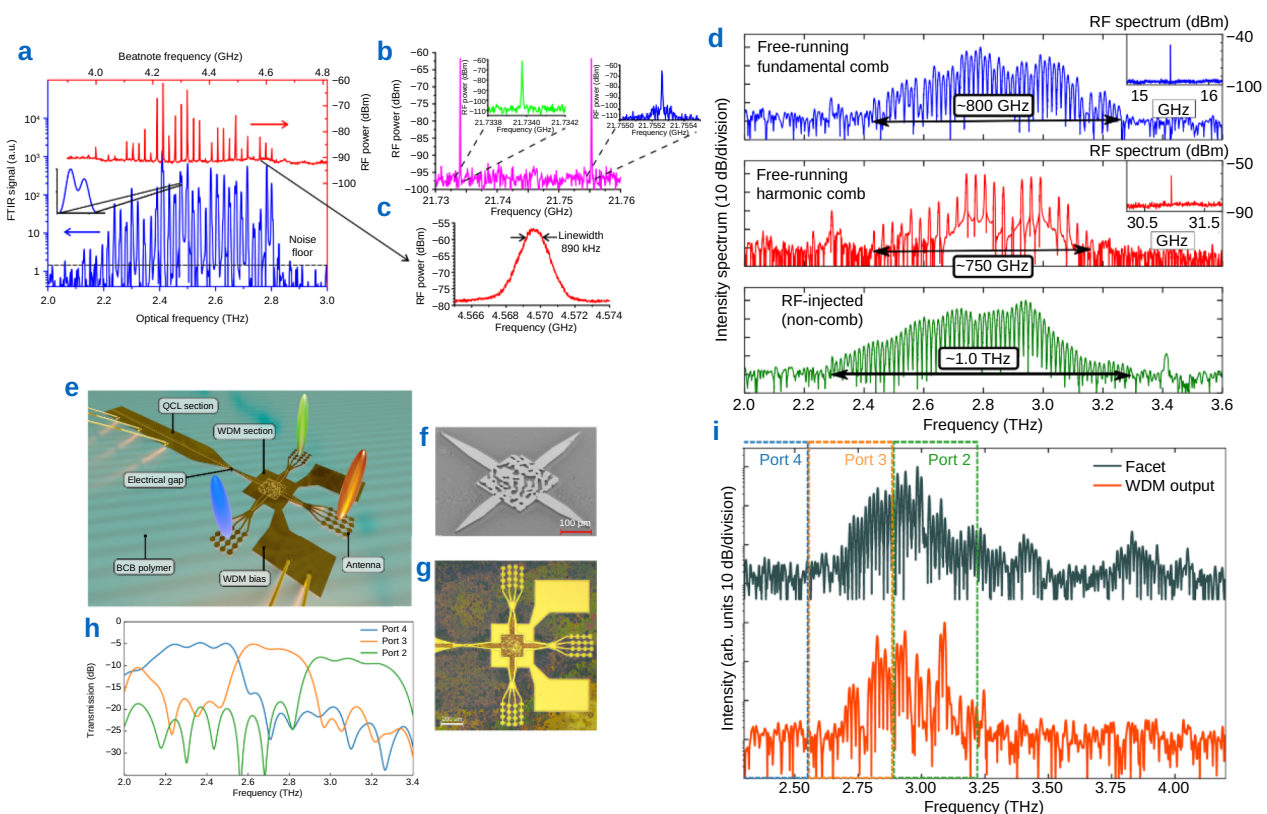


Fig. 18 | Integrated THz QCL DCS. (a) Optical spectrum (blue curve) of the dual-comb measured with a Bruker 80v FTIR. The inset shows that the modes consist of two peaks separated by 4.4 GHz close to the resolution limit of the FTIR (2.25 GHz) corresponding to the two combs. In red is the corresponding multi-heterodyne spectrum extracted from the current bias of the LO laser. (b) Corresponding intermode beatnote signal. The insets show zooms of the individual beatnotes showing linewidths of below 1 kHz. (c) Zoom of one line of the multi-heterodyne spectrum. (d) Measured THz and RF spectrum of the 10% reflectivity inverse-designed facet device in CW at a heat sink temperature of 30 K. (e) Illustration of the planarized device structure with the active material surrounded by BCB. (f) SEM image of the dryetched WDM inverse-designed region before planarization. (g) Optical microscope image of a fully fabricated device. (h) 3D CST numerical simulation results for the transmission of the QCL section output to the WDM output ports. (i) Spectral measurements of the QCL and WDM outputs. Figure reproduced with permission from: (a–c) ref. ¹¹², (d) ref. ¹¹⁵, (e–i) ref. ¹¹⁶, under a Creative Commons Attribution License.

on-chip dual-comb systems are compact, their application in real substance detection is hindered by structural limitations. In double-metal waveguide QCLs¹¹³, optical coupling between the combs occurs via the edge of the bottom metal, whereas in single plasmon waveguide QCLs¹¹⁴, it happens through the laser substrate. As a result, an on-chip configuration does not allow for the placement of a sample between the two combs. Furthermore, they present a planarized THz QCL with an inverse-designed end facet reflector coupled to a surface-emitting patch array antenna, and demonstrated a planar THz QCL frequency comb covering 800 GHz with peak power as high as 13.5 mW, as shown in Fig. 18(d)¹¹⁵. Recently, the team demonstrated an on-chip system integrating an active three-channel wavelength division multiplexer (WDM) and equipped with a THz QCL comb. The WDM input is coupled to an integrated THz QCL frequency comb, providing three 330 GHz broadband output ports, ranging from 2.2–3.2 THz, with a maximum crosstalk of –6 dB, as shown in Fig. 18(e–i)¹¹⁶.

In 2019, Li et al. developed a compact THz multiheterodyne spectrometer utilizing two spatially separated QCL frequency combs, which allows rapid spectral detection by positioning samples between them¹¹⁷. This setup successfully established a reliable microwave-to-THz frequency link, as demonstrated by DCS measured at varying relative humidity (RH) levels. Further, they developed several approaches to achieve stable THz QCL DCS sources^{118,119}. In 2023, Li et al.¹²⁰ proposed a self-referenced frequency stabilization method for THz-QCL dual-comb systems, eliminating reliance on complex external phase-locked electronic equipment. This approach effectively suppresses common-mode noise in the dual-comb carrier frequency and significantly reduces repetition frequency noise by filtering one dual-comb spectral line and mixing it with the entire dual-comb signal. Experimental results demonstrate that the self-referenced dual-comb signal achieves a “maximum hold” linewidth of 14.8 kHz over 60 seconds, representing over 130-fold improvement in long-term stability compared to the 2 MHz (15 seconds) unstabilized performance, as shown in (Fig. 19(a, b)). This work provides a concise and effective technical pathway for realizing highly stable, all-solid-state THz-DCS systems, suitable for high-precision spectroscopy and metrology applications.

Further advancing the field, in 2025, Liu¹²¹ successfully demonstrated a compact THz triple-comb source based on three THz-QCLs operating at approximately 4.2 THz. Through finite element simulations, the study revealed the optical interaction mechanism between lasers, primarily substrate coupling. Leveraging the QCL's inherent rapid self-detection capability, simultaneous measurement of multiple dual-comb signals was achieved. This allows for the acquisition of richer spectroscopic information in a single experiment, effectively reducing systematic errors and enhancing measurement accuracy. The experimentally measured three-comb signals exhibit a signal-to-noise ratio exceeding 30 dB

and possess sufficient comb teeth to support spectroscopy measurements (Fig. 19(c, d)). This approach provides a novel multi-comb solution for high-precision spectroscopy and metrology applications in the THz band, eliminating the need for mechanical moving parts.

In addition to comb stabilization, THz-QCL systems face several challenges. One major limitation arises from the scarcity of high-sensitivity, broadband, and fast THz detectors. Compared with mature mid-infrared photodetection technologies, THz detection often requires cryogenic operation or suffers from reduced responsivity and bandwidth, thereby limiting the achievable signal-to-noise ratio and acquisition speed in dual-comb spectroscopy. Another critical issue is thermal management. Owing to the strong temperature dependence of intersubband transitions in THz-QCLs, excess heat accumulation can alter dispersion characteristics and degrade comb coherence. Although substantial progress has been achieved through advanced heat-sinking strategies, optimized waveguide designs, and cryogenic packaging, thermal effects remain a key obstacle toward high-power, broadband, and room-temperature THz-QCL dual-comb systems.

Advances in achieving metrology-grade performance have driven the development of broadband THz combs (Fig. 20). These combs have emerged as critical tools across multiple scientific and technological frontiers. Serving as THz frequency standards, they enable the stabilization of THz QCLs and the generation of high-power, stable THz waves, with promising applications in high-precision radar, next-generation wireless communications, and industrial non-destructive testing. Their broad spectral coverage and high stability allow for the simultaneous detection of multiple non-adjacent molecular spectral lines, providing key tools for fundamental research—from molecular structure analysis to the measurement of fundamental physical constants. Furthermore, their enhanced resolution and accuracy benefit a wide array of sensing applications, including trace gas detection, environmental monitoring, and medical breath diagnostics. With the continuous optimization of source performance and ongoing innovation in system architecture, THz dual-comb spectroscopy technology is steadily advancing toward higher precision, enhanced stability, and broader spectral coverage. This progress is establishing a robust technical foundation for the deepening exploration of THz science and the expansion of its practical applications.

5 Challenges and outlooks

The preceding sections have reviewed mid-infrared and THz DCS separately from the perspective of source technologies and representative demonstrations. However, many of the remaining bottlenecks are shared at the system level. In both spectral domains, practical progress depends not only on source generation, but also on how broadband spectral

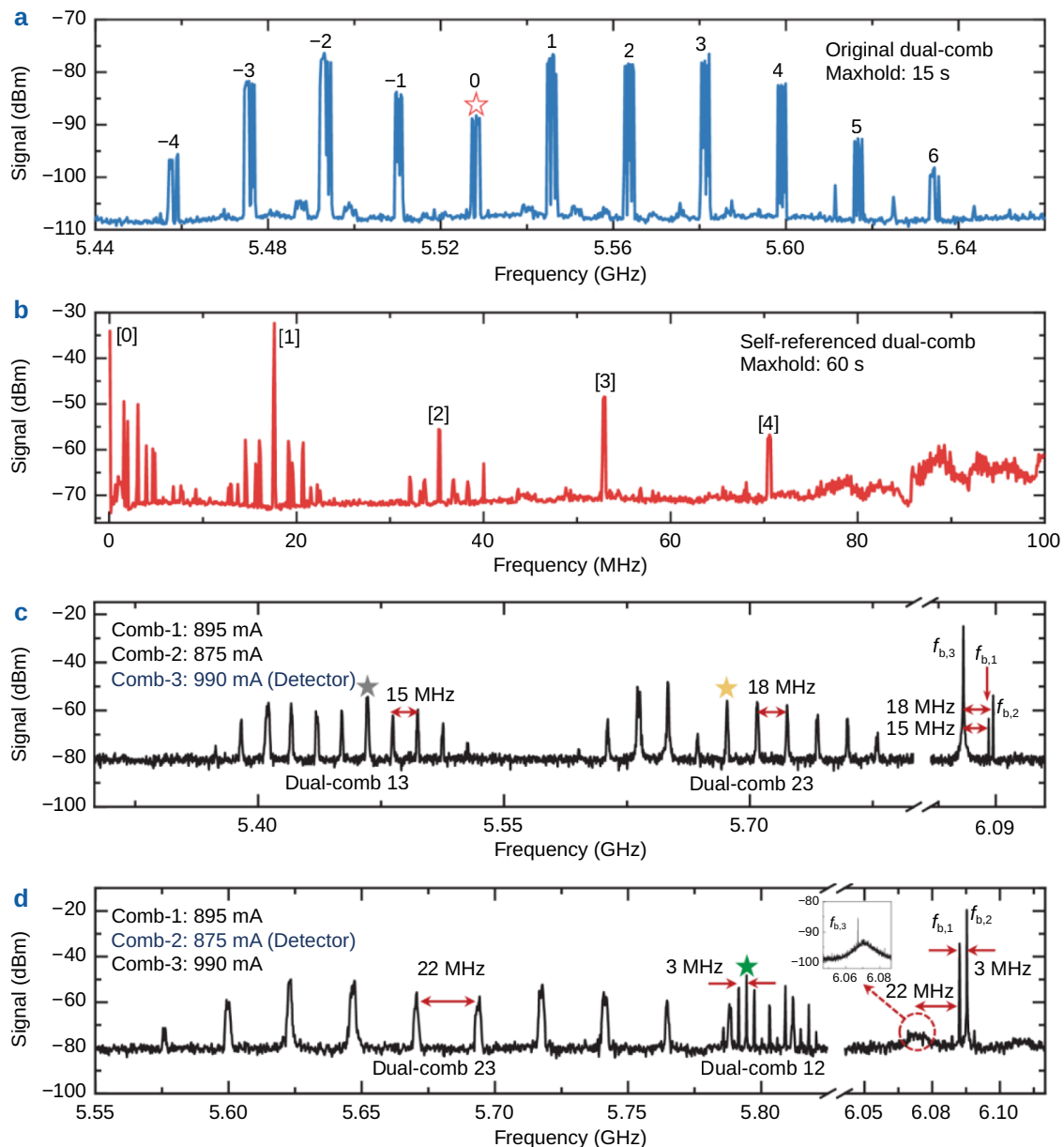


Fig. 19 | THz-QCL dual-comb spectroscopy. (a) Raw dual-comb spectrum acquired without the self-referencing method, using a 15-second max-hold measurement. The spectral line labeled “0” and marked with a red asterisk is the selected comb tooth used for self-referencing and mixing. (b) Self-referenced dual-comb spectrum acquired using a 60-second max-hold measurement. For both (a) and (b), the resolution bandwidth (RBW) and video bandwidth (VBW) were set to 500 Hz and 50 Hz, respectively. (c) Multi-heterodyne triple-comb signals and intermode beat notes for dual-comb 13 and dual-comb 23, measured with comb 3 serving as the fast detector. (d) Multi-heterodyne triple-comb signals and intermode beat notes for dual-comb 23 and dual-comb 12, measured with comb 2 as the detector. The inset displays the narrow-linewidth signals acquired with RBW = 500 Hz and VBW = 50 Hz. Figure reproduced with permission from: (a, b) ref.¹²⁰, (c, d) ref.¹²¹, under a Creative Commons Attribution License.

coverage, sensitivity preservation, detection architecture, and long-term coherence control can be jointly optimized. From this perspective, the following discussion shifts from wavelength-specific implementations to the common challenges and complementary opportunities that connect mid-infrared and THz DCS.

5.1 Toward full infrared-to-THz spectral coverage

Despite the remarkable progress of DCS in both the mid-infrared and THz regions, achieving continuous infrared-to-THz spectral coverage remains a major challenge, especially across the 15–60 μm (5–20 THz) band. In

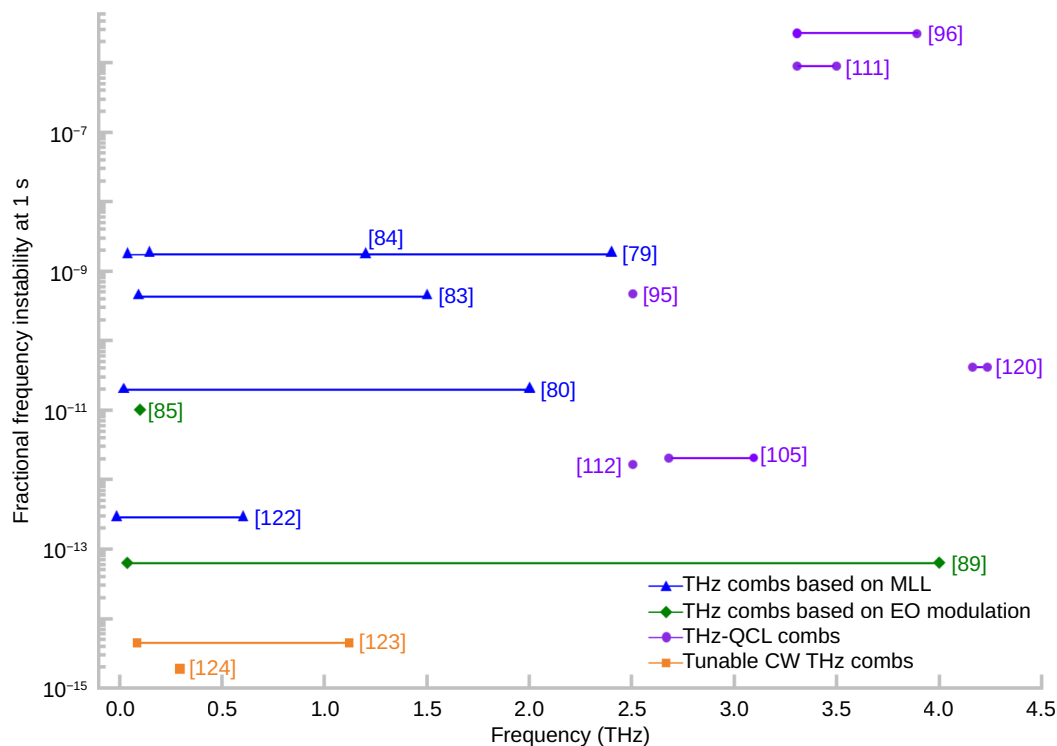


Fig. 20 | Comparison of precision and spectral width for different THz frequency combs. Data points include: THz combs based on MLL (blue triangles)^{79,80,83,84,122}; THz combs based on EO modulation (green diamonds)^{85,89}; THz-QCL combs (purple circles)^{95,96,105,111,112,120}; and tunable CW THz combs via photomixing of two CW lasers (yellow squares)^{123,124}.

practice, the currently accessible spectral windows remain strongly platform dependent. IP-DFG based on multiple $\chi^{(2)}$ crystals has already extended mid-infrared comb coverage to 3–13 μm , while coherent long-wave mid-infrared sources based on longer-wavelength nonlinear materials have further pushed the accessible range to about 18 μm . At the same time, direct-emission THz QCL combs and QCL-based DFG have opened the low-THz region, particularly around 1–5 THz. However, these advances still do not yield continuous infrared-to-THz coverage, because operation toward higher THz frequencies is increasingly constrained by material absorption, waveguide loss, and the Reststrahlen band of typical III-V semiconductors¹²⁵.

Therefore, a more realistic route toward full-band fingerprint spectroscopy is likely to rely on segmented spectral engineering, in which multiple nonlinear crystals and complementary source architectures are combined to cover different spectral windows. Future progress will likely come from broader long-wave mid-infrared generation using optimized $\chi^{(2)}$ materials, continued advances in QCL-based comb sources toward and within the THz region¹²⁶, and hybrid conversion architectures that coherently connect these otherwise separated spectral regimes^{127,128}.

Beyond spectral coverage, beam quality and mode matching strongly affect how efficiently comb power is delivered and utilized in practical DCS systems. Parameters such as M^2 , divergence, spatial-mode quality, and pointing stability

influence coupling efficiency, throughput, and baseline stability in cavities, multipass cells, waveguides, open-path links, and imaging setups. DFG- and OPO-based combs generally inherit good spatial modes from near-infrared pumps, whereas QCL- and THz-QCL-based combs often require beam shaping due to divergent or astigmatic outputs. For integrated platforms, efficient mode matching between on-chip and external optics is equally important. Although beam-quality metrics are not yet consistently reported across DCS studies, they should be considered alongside spectral coverage, coherence, output power, and detector performance when selecting a DCS architecture.

Another key issue is broadband detection across the infrared-to-THz range. In practice, no single detector technology is optimal over all relevant wavelengths. In the mid-infrared, detector noise, speed, and saturation limitations have motivated heterogeneous readout strategies such as nonlinear up-conversion and cross-comb detection, which transfer the spectral information to the near-infrared for low-noise photodetection. Notably, EO sampling has recently enabled instantaneous octave-wide dual-comb coverage from the mid-infrared to the THz range, while QCL-based self-detection has shown that one comb can simultaneously function as emitter, local oscillator, and detector. This idea has progressed from on-chip THz self-detected DCS to compact THz QCL self-detection, and more recently to self-detecting mid-infrared QCL dual-comb

systems. These developments suggest that one solution for future full-band DCS will be a combination of heterogeneous or self-detecting readout strategies tailored to different spectral regions.

In parallel, a variety of indirect measurement methods have also been proposed. By converting molecular absorption into acoustic, thermal, or mechanical signals, these methods reduce the dependence on wavelength-specific photodetectors and can, in principle, be applied across a wide range of spectral regions. For example, photo-acoustic DCS was experimentally demonstrated by directly detecting the pressure waves generated by dual-comb excitation to reconstruct broadband molecular spectra¹²⁹.

Notably, indirect DCS approaches, such as quartz-enhanced, cavity-enhanced, and cantilever-enhanced photo-acoustic DCS, have enabled high sensitivity for molecular fingerprinting. For instance, cavity-enhanced photo-acoustic DCS reported a minimum detection limit of 0.6 ppb for acetylene at an integration time of 100 s¹³⁰. A closely related route is photo-thermal DCS, in which molecular absorption is converted into refractive-index or volume changes. This method achieved tooth-resolved acetylene spectroscopy over more than 1 THz, with a minimum detection limit of 8.7 ppm³². Moreover, dual-comb optomechanical spectroscopy was demonstrated using a high-Q nanomechanical membrane inside an optical cavity as the transducer³³. This approach enabled broadband acetylene spectroscopy over more than 2 THz at the ppb level, with a normalized noise-equivalent absorption coefficient of $1.71 \times 10^{-11} \text{ cm}^{-1} \cdot \text{W} \cdot \text{Hz}^{-1/2}$.

In short, realizing full infrared-to-THz DCS will critically depend on overcoming two closely connected challenges: the generation of broadband comb sources across otherwise separated spectral windows, and the implementation of effective detection or transduction schemes that preserve sensitivity across those windows. In this sense, full-band DCS is not a source problem alone, but a system-level problem involving coordinated advances in source engineering, readout strategy, and sensitivity preservation.

5.2 Towards quantum-enhanced DCS

As the spectral bandwidth broadens, the power available per comb tooth decreases, leading to a lower signal-to-noise ratio in broadband DCS. To address this limitation, a variety of enhancement strategies, including cavity-enhanced DCS and related approaches, have been developed and have brought DCS sensitivity close to the shot-noise limit. Consequently, overcoming this quantum-noise boundary has become an important direction for the future development of DCS.

In recent years, quantum technologies (such as squeezing and entanglement) have provided new possibilities for surpassing the classical sensitivity limit^{131,132}. A squeezed-state DCS has been experimentally demonstrated, utilizing a

nonlinear Mach-Zehnder interferometer to generate an amplitude-squeezed quantum OFC¹³³. This squeezed comb was then combined with a coherent-state comb for hydrogen sulfide (H_2S) absorption spectrum measurement, with measurement residuals approximately 2.9 dB below the shot noise limit. To extend the spectral range into the mid-infrared, a quantum correlation-enhanced DCS combined with frequency upconversion has been proposed¹³⁴. By compressing the intensity difference noise between a pair of twin-correlated OFCs, quantum-enhanced mid-infrared DCS has been realized, with signal-to-noise ratios exceeding the classical limit by approximately 2 dB. Based on the theoretical foundation in the literature¹³⁵, customized entanglement structures in the frequency comb were utilized to simultaneously detect all comb lines below the classical DCS standard quantum limit¹³⁶. When applied to gas detection, the SNR was improved by 2.6 dB, and the integration time was reduced by 1.7 times compared to classical DCS.

5.3 Coherence preservation and long-term stability

For DCS to become truly viable for field-deployable applications, environmental robustness must be significantly improved, because practical operation requires long averaging times to reach high sensitivity. However, in DCS, the sensitivity gain obtained through longer averaging is fundamentally limited by the mutual coherence time of the two combs. Under practical conditions, environmental perturbations such as temperature fluctuations, mechanical vibrations, and air-path instability can further shorten this coherence time and degrade measurement reliability. These constraints have driven a clear methodological evolution in coherence recovery, from passive schemes relying on the intrinsic coherence of the sources, to active control referenced to external standards, and more recently to data-driven digital correction strategies. Although the specific implementations differ between mid-infrared and THz systems, the underlying challenge is the same: practical sensitivity gains require longer coherent averaging, which in turn demands increasingly robust mutual coherence under realistic environmental perturbations.

In the passive regime, coherence is preserved mainly through common-mode source design, so that the two combs share as much optical or electronic origin as possible. This approach is attractive because it greatly simplifies the system and can already provide excellent mutual coherence; for example, passive coherent dual-comb spectroscopy based on optical-optical modulation achieved a relative comb-tooth linewidth of 10 mHz and a coherence time of 100 s without fast phase-locking electronics or post phase correction¹³⁷. However, because passive schemes suppress mainly common-mode fluctuations, they remain sensitive to residual differential noise from external modulation sources, non-common-path perturbations, and long-term environmental drifts.

To suppress these residual fluctuations, active control referenced to external standards was developed. In the mid-infrared, feed-forward dual-comb spectroscopy has demonstrated experimental coherence times exceeding half an hour, while more practical active-control implementations have combined adaptive sampling with simple frequency stabilization to realize continuous real-time operation over 1200 h¹³⁸; in the THz regime, an all-fiber adaptive clock together with a real-time feedback loop has further enabled narrow-linewidth long-term-stable dual-comb operation⁸⁴. These active approaches, however, come at the cost of greater hardware complexity and more stringent reference requirements.

To reduce this burden, coherence recovery has increasingly been extended into the data domain, where the key task is to extract, estimate, and compensate the relative phase and timing fluctuations between the two combs. In practice, this is typically achieved through error-signal retrieval, phase correction, and numerical resampling, allowing coherent reconstruction of the interferograms. By moving beyond the bandwidth limitations of conventional closed-loop control, these methods can significantly extend the effective coherent averaging time and support long-term, high-precision gas spectroscopy^{139,140}. Early approaches often relied on external references or auxiliary error-measurement channels, and were therefore vulnerable to mismatches between the reference path and the main interferometric acquisition path¹⁴¹. More recently, self-referenced correction methods based directly on the interferograms themselves have been developed¹⁴², reducing the reliance on additional reference channels.

6 Summary

In summary, this review highlights recent advances in mid-infrared and THz DCS, a technique renowned for high spectral resolution, rapid acquisition, and intrinsic frequency accuracy without moving parts. We examine key methods for generating dual-comb sources—including QCLs, OPOs, DFG, and microresonator-based combs—analyzing their performance, trade-offs, and recent innovations. The review also addresses ongoing challenges, such as broadband spectral coverage, high-sensitivity detection, long-term stability and looks forward to future directions involving integrated, field-deployable platforms, and quantum-enhanced techniques. Looking ahead, these developments are expected to further drive mid-infrared and terahertz DCS from laboratory demonstrations toward broader practical deployment in trace-gas sensing, oil-and-gas leak detection, atmospheric and environmental monitoring, biomedical diagnostics, and precision molecular spectroscopy, while also opening new opportunities in dual-comb ranging and spectroscopic imaging.

References

- Fortier T, Baumann E. 20 years of developments in optical frequency comb technology and applications. *Commun Phys* 2, 153 (2020).
- Diddams SA, Vahala K, Udem T. Optical frequency combs: coherently uniting the electromagnetic spectrum. *Science* 369, eaay3676 (2020).
- Hall JL. Nobel lecture: defining and measuring optical frequencies. *Rev Mod Phys* 78, 1279–1295 (2006).
- Diddams SA, Hollberg L, Mbele V. Molecular fingerprinting with the resolved modes of a femtosecond laser frequency comb. *Nature* 445, 627–630 (2007).
- Picqué N, Hänsch TW. Frequency comb spectroscopy. *Nat Photonics* 13, 146–157 (2019).
- Rieker GB, Giorgetta FR, Swann WC et al. Frequency-comb-based remote sensing of greenhouse gases over kilometer air paths. *Optica* 1, 290–298 (2014).
- Zhang CK, Ooi T, Higgins JS et al. Frequency ratio of the ^{229m}Th nuclear isomeric transition and the ⁸⁷Sr atomic clock. *Nature* 633, 63–70 (2024).
- Adler F, Maslowski P, Foltynowicz A et al. Mid-infrared Fourier transform spectroscopy with a broadband frequency comb. *Opt Express* 18, 21861–21872 (2010).
- Grillii R, Méjean G, Kassi S et al. Frequency comb based spectrometer for *in situ* and real time measurements of IO, BrO, NO₂, and H₂CO at PPTV and PPQV levels. *Environ Sci Technol* 46, 10704–10710 (2012).
- Yasui T, Saneyoshi E, Araki T. Asynchronous optical sampling terahertz time-domain spectroscopy for ultrahigh spectral resolution and rapid data acquisition. *Appl Phys Lett* 87, 061101 (2005).
- Coddington I, Swann WC, Newbury NR. Time-domain spectroscopy of molecular free-induction decay in the infrared. *Opt Lett* 35, 1395–1397 (2010).
- Millot G, Pitois S, Yan M et al. Frequency-agile dual-comb spectroscopy. *Nat Photonics* 10, 27–30 (2016).
- Long DA, Cich MJ, Mathurin C et al. Nanosecond time-resolved dual-comb absorption spectroscopy. *Nat Photonics* 18, 127–131 (2024).
- Coluccelli N, Cassinerio M, Redding B et al. The optical frequency comb fibre spectrometer. *Nat Commun* 7, 12995 (2016).
- Mandon J, Guelachvili G, Picqué N. Fourier transform spectroscopy with a laser frequency comb. *Nat Photonics* 3, 99–102 (2009).
- Maslowski P, Lee KF, Johansson AC et al. Surpassing the path-limited resolution of Fourier-transform spectrometry with frequency combs. *Phys Rev A* 93, 021802(R) (2016).
- Long DA, Fleisher AJ, Plusquellic DF et al. Multiplexed sub-Doppler spectroscopy with an optical frequency comb. *Phys Rev A* 94, 061801(R) (2016).
- Meek SA, Hipke A, Guelachvili G et al. Doppler-free Fourier transform spectroscopy. *Opt Lett* 43, 162–165 (2018).
- Trocha P, Karpov M, Ganin D et al. Ultrafast optical ranging using microresonator soliton frequency combs. *Science* 359, 887–891 (2018).
- Dutt A, Joshi C, Ji XC et al. On-chip dual-comb source for spectroscopy. *Sci Adv* 4, e1701858 (2018).
- Giorgetta FR, Potvin S, Deschênes JD et al. Free-form dual-comb spectroscopy for compressive sensing and imaging. *Nat Photonics* 18, 1312–1319 (2024).
- Dudley JM, Genty G, Coen S. Supercontinuum generation in photonic crystal fiber. *Rev Mod Phys* 78, 1135–1184 (2006).
- Lind AJ, Kowligy A, Timmers H et al. Mid-infrared frequency comb generation and spectroscopy with few-cycle pulses and $\chi^{(2)}$ non-linear optics. *Phys Rev Lett* 124, 133904 (2020).
- Lesko DMB, Timmers H, Xing SD et al. A six-octave optical

- frequency comb from a scalable few-cycle erbium fibre laser. *Nat Photonics* **15**, 281–286 (2021).
25. Xu BX, Chen ZJ, Hänsch TW et al. Near-ultraviolet photon-counting dual-comb spectroscopy. *Nature* **627**, 289–294 (2024).
 26. Chen J, Nitta K, Zhao X et al. Adaptive-sampling near-Doppler-limited terahertz dual-comb spectroscopy with a free-running single-cavity fiber laser. *Adv Photonics* **2**, 036004 (2020).
 27. Hoghooghi N, Wright RJ, Makowiecki AS et al. Broadband coherent cavity-enhanced dual-comb spectroscopy. *Optica* **6**, 28–33 (2019).
 28. Xu K, Zhao X, Wang Z et al. Multipass-assisted dual-comb gas sensor for multi-species detection using a free-running fiber laser. *Appl Phys B* **126**, 39 (2020).
 29. Tomaszewska-Rolla D, Jaworski P, Wu DK et al. Mid-infrared optical frequency comb spectroscopy using an all-silica antiresonant hollow-core fiber. *Opt Express* **32**, 10679–10689 (2024).
 30. Geng XT, Chun BJ, Seo JH et al. Frequency comb transferred by surface plasmon resonance. *Nat Commun* **7**, 10685 (2016).
 31. Friedlein JT, Baumann E, Briggman KA et al. Dual-comb photoacoustic spectroscopy. *Nat Commun* **11**, 3152 (2020).
 32. Wang Q, Wang Z, Zhang H et al. Dual-comb photothermal spectroscopy. *Nat Commun* **13**, 2181 (2022).
 33. Ren XY, Pan J, Yan M et al. Dual-comb optomechanical spectroscopy. *Nat Commun* **14**, 5037 (2023).
 34. Coddington I, Newbury N, Swann W. Dual-comb spectroscopy. *Optica* **3**, 414–426 (2016).
 35. Schliesser A, Picqué N, Hänsch TW. Mid-infrared frequency combs. *Nat Photonics* **6**, 440–449 (2012).
 36. Tonouchi M. Cutting-edge terahertz technology. *Nat Photonics* **1**, 97–105 (2007).
 37. Fellinger J, Winkler G, Aldia PEC et al. Simple approach for extending the ambiguity-free range of dual-comb ranging. *Opt Lett* **46**, 3677–3680 (2021).
 38. Cruz FC, Maser DL, Johnson T et al. Mid-infrared optical frequency combs based on difference frequency generation for molecular spectroscopy. *Opt Express* **23**, 26814–26824 (2015).
 39. Baumann E, Giorgetta FR, Swann WC et al. Spectroscopy of the methane ν_3 band with an accurate midinfrared coherent dual-comb spectrometer. *Phys Rev A* **84**, 062513 (2011).
 40. Chen ZJ, Hänsch TW, Picqué N. Mid-infrared feed-forward dual-comb spectroscopy. *Proc Natl Acad Sci USA* **116**, 3454–3459 (2019).
 41. Parriaux A, Hammani K, Millot G. Electro-optic frequency combs. *Adv Opt Photonics* **12**, 223–287 (2020).
 42. Yan M, Luo PL, Iwakuni K et al. Mid-infrared dual-comb spectroscopy with electro-optic modulators. *Light Sci Appl* **6**, e17076 (2017).
 43. Nitzsche L, Goldschmidt J, Kiessling J et al. Tunable dual-comb spectrometer for mid-infrared trace gas analysis from 3 to 4.7 μm . *Opt Express* **29**, 25449–25461 (2021).
 44. Yuan ZC, Fan XY, Xu BX et al. Digitally generated high-resolution mid-infrared dual-comb spectroscopy system based on electro-optic modulation. *Opt Lett* **49**, 5711–5714 (2024).
 45. Luo PL. Long-wave mid-infrared time-resolved dual-comb spectroscopy of short-lived intermediates. *Opt Lett* **45**, 6791–6794 (2020).
 46. Liu MC, Gray RM, Costa L et al. Mid-infrared cross-comb spectroscopy. *Nat Commun* **14**, 1044 (2023).
 47. Ycas G, Giorgetta FR, Baumann E et al. High-coherence mid-infrared dual-comb spectroscopy spanning 2.6 to 5.2 μm . *Nat Photonics* **12**, 202–208 (2018).
 48. Hoghooghi N, Xing SD, Chang P et al. Broadband 1-GHz mid-infrared frequency comb. *Light Sci Appl* **11**, 264 (2022).
 49. Vasilyev S, Muraviev A, Konnov D et al. Longwave infrared (6.6–11.4 μm) dual-comb spectroscopy with 240,000 comb-mode-resolved data points at video rate. *Opt Lett* **48**, 2273–2276 (2023).
 50. Zhang ZW, Gardiner T, Reid DT. Mid-infrared dual-comb spectroscopy with an optical parametric oscillator. *Opt Lett* **38**, 3148–3150 (2013).
 51. Muraviev AV, Smolski VO, Loparo ZE et al. Massively parallel sensing of trace molecules and their isotopologues with broadband subharmonic mid-infrared frequency combs. *Nat Photonics* **12**, 209–214 (2018).
 52. Kara O, Zhang Z, Gardiner T et al. Dual-comb mid-infrared spectroscopy with free-running oscillators and absolute optical calibration from a radio-frequency reference. *Opt Express* **25**, 16072–16082 (2017).
 53. Abbas MA, Pan Q, Mandon J et al. Time-resolved mid-infrared dual-comb spectroscopy. *Sci Rep* **9**, 17247 (2019).
 54. Bauer CP, Camenzind SL, Pupeikis J et al. Dual-comb optical parametric oscillator in the mid-infrared based on a single free-running cavity. *Opt Express* **30**, 19904–19921 (2022).
 55. Bauer CP, Bejm ZA, Bollier MK et al. High-sensitivity dual-comb and cross-comb spectroscopy across the infrared using a widely tunable and free-running optical parametric oscillator. *Nat Commun* **15**, 7211 (2024).
 56. Bauer CP, Lang L, Willenberg B et al. Ultrafast 1-GHz repetition rate dual-comb optical parametric oscillator. *Opt Express* **33**, 39130–39139 (2025).
 57. Hugi A, Villares G, Blaser S et al. Mid-infrared frequency comb based on a quantum cascade laser. *Nature* **492**, 229–233 (2012).
 58. Villares G, Hugi A, Blaser S et al. Dual-comb spectroscopy based on quantum-cascade-laser frequency combs. *Nat Commun* **5**, 5192 (2014).
 59. Villares G, Wolf J, Kazakov D et al. On-chip dual-comb based on quantum cascade laser frequency combs. *Appl Phys Lett* **107**, 251104 (2015).
 60. Hillbrand J, Andrews AM, Detz H et al. Coherent injection locking of quantum cascade laser frequency combs. *Nat Photonics* **13**, 101–104 (2019).
 61. Teng CC, Chen YF, Liu J et al. Frequency stabilization and referencing of a mid-infrared dual-comb spectrometer. *Appl Phys B* **132**, 23 (2026).
 62. Hillbrand J, Bertrand M, Wittwer V et al. Synchronization of frequency combs by optical injection. *Opt Express* **30**, 36087–36095 (2022).
 63. Jouy P, Wolf JM, Bidaux Y et al. Dual comb operation of $\lambda \sim 8.2 \mu\text{m}$ quantum cascade laser frequency comb with 1 W optical power. *Appl Phys Lett* **111**, 141102 (2017).
 64. Lu QY, Manna S, Wu DH et al. Shortwave quantum cascade laser frequency comb for multi-heterodyne spectroscopy. *Appl Phys Lett* **112**, 141104 (2018).
 65. Pinkowski NH, Cassidy SJ, Strand CL et al. Quantum-cascade-laser-based dual-comb thermometry and speciation at high temperatures. *Meas Sci Technol* **32**, 035501 (2021).
 66. Gianella M, Nataraj A, Tuzson B et al. High-resolution and gapless dual comb spectroscopy with current-tuned quantum cascade lasers. *Opt Express* **28**, 6197–6208 (2020).
 67. Lepère M, Browet O, Clément J et al. A mid-infrared dual-comb spectrometer in step-sweep mode for high-resolution molecular spectroscopy. *J Quant Spectrosc Radiat Transf* **287**, 108239 (2022).
 68. Huang RX, Ma Y, Wu DP et al. Longwave infrared dual-comb spectroscopy based on high power quantum cascade lasers. *Infrared Phys Technol* **150**, 105965 (2025).
 69. Hayden J, Geiser M, Gianella M et al. Mid-infrared dual-comb spectroscopy with quantum cascade lasers. *APL Photonics* **9**, 031101 (2024).
 70. Chang L, Liu ST, Bowers JE. Integrated optical frequency comb technologies. *Nat Photonics* **16**, 95–108 (2022).
 71. Yu MJ, Okawachi Y, Griffith AG et al. Silicon-chip-based mid-infrared dual-comb spectroscopy. *Nat Commun* **9**, 1869 (2018).

72. Guo HR, Weng WL, Liu JQ et al. Nanophotonic supercontinuum-based mid-infrared dual-comb spectroscopy. *Optica* 7, 1181–1188 (2020).
73. Bao CY, Yuan ZQ, Wu L et al. Architecture for microcomb-based GHz-mid-infrared dual-comb spectroscopy. *Nat Commun* 12, 6573 (2021).
74. Yoshii K, Kuse N, Inoue K et al. Generation of a mW-class broadband mid-infrared comb using a waveguide-type PPLN crystal and its application to dual-comb spectroscopy. In *2022 Conference on Lasers and Electro-Optics Pacific Rim 1–2* (IEEE, 2022). <http://doi.org/10.1109/CLEO-PR62338.2022.10432633>.
75. Zhou L, Lou HP, Deng ZJ et al. Compact mid-infrared dual-comb spectrometer over 3–4 μm via intra-pulse difference frequency generation in LiNbO_3 waveguides. *High Power Laser Sci Eng* 12, e23 (2024).
76. Fu BT, Gao RH, Yao N et al. Soliton microcomb generation by cavity polygon modes. *Opto-Electron Adv* 7, 240061 (2024).
77. Liu ML, Wei ZQ, Zhu HT et al. Soliton microcombs in optical microresonators with perfect spectral envelopes. *Opto-Electron Adv* 8, 240257 (2025).
78. Yasui T, Kabetani Y, Saneyoshi E et al. Terahertz frequency comb by multifrequency-heterodyning photoconductive detection for high-accuracy, high-resolution terahertz spectroscopy. *Appl Phys Lett* 88, 241104 (2006).
79. Finneran IA, Good JT, Holland DB et al. Decade-spanning high-precision terahertz frequency comb. *Phys Rev Lett* 114, 163902 (2015).
80. Hsieh YD, Lyonaga Y, Sakaguchi Y et al. Spectrally interleaved, comb-mode-resolved spectroscopy using swept dual terahertz combs. *Sci Rep* 4, 3816 (2014).
81. Konnov D, Muraviev A, Vasilyev S et al. High-resolution frequency-comb spectroscopy with electro-optic sampling and instantaneous octave-wide coverage across mid-IR to THz at a video rate. *APL Photonics* 8, 110801 (2023).
82. Hu GQ, Mizuguchi T, Oe R et al. Dual terahertz comb spectroscopy with a single free-running fibre laser. *Sci Rep* 8, 11155 (2018).
83. Nakajima Y, Yumoto T, Uchiyama R et al. Mechanical-sharing dual-comb fiber laser for terahertz dual-comb spectroscopy. In *CLEO: Science and Innovations 2024 SF3N. 7* (Optica Publishing Group, 2024). https://doi.org/10.1364/CLEO_SI.2024.SF3N.7.
84. Xia Y, Li M, Liu Z et al. A narrow linewidth terahertz dual-comb spectrometer with an all-fiber adaptive clock and a real-time feedback loop. *Opt Laser Technol* 162, 109314 (2023).
85. Jerez B, Walla F, Betancur A et al. Electro-optic THz dual-comb architecture for high-resolution, absolute spectroscopy. *Opt Lett* 44, 415–418 (2019).
86. Fang X, Zhang L, Yang ZM et al. Photonic terahertz hyperspectral imaging with swept-frequency dual combs. *Laser Photonics Rev* 19, 2400696 (2025).
87. Stroud JR, Plusquellic DF. Dual chirped-pulse electro-optical frequency comb method for simultaneous molecular spectroscopy and dynamics studies: formic acid in the terahertz region. *Opt Lett* 47, 3716–3719 (2022).
88. Abdollahi S, Ladouce M, Marin-Palomo P et al. Agile THz-range spectral multiplication of frequency combs using a multi-wavelength laser. *Nat Commun* 15, 1305 (2024).
89. Li M, Xia Y, Chen Y et al. Broadband terahertz comb with sub-Hz comb linewidth. *Adv Photonics* 8, 026015 (2026).
90. Kazarinov RF, Suris RA. Possible amplification of electromagnetic waves in a semiconductor with a superlattice. *Sov Phys Semicond* 5, 707–709 (1971).
91. Faist J, Capasso F, Sivco DL et al. Quantum cascade laser. *Science* 264, 553–556 (1994).
92. Köhler R, Tredicucci A, Beltram F et al. Terahertz semiconductor-heterostructure laser. *Nature* 417, 156–159 (2002).
93. Heckelmann I, Bertrand M, Dikopoltsev A et al. Quantum walk comb in a fast gain laser. *Science* 382, 434–438 (2023).
94. Liu BB, Silvestri C, Zhou K et al. Terahertz semiconductor laser chaos. *Nat Commun* 16, 9985 (2025).
95. Rösch M, Scalari G, Beck M et al. Octave-spanning semiconductor laser. *Nat Photonics* 9, 42–47 (2015).
96. Zhou K, Liu BB, Wan WJ et al. Microwave injection induced broadband dual-comb operation in terahertz quantum cascade lasers with a stacked active region. *ACS Photonics* 12, 747–757 (2025).
97. Burghoff D, Kao TY, Han NR et al. Terahertz laser frequency combs. *Nat Photonics* 8, 462–467 (2014).
98. Wang FH, Nong H, Fobbe T et al. Short terahertz pulse generation from a dispersion compensated modelocked semiconductor laser. *Laser Photonics Rev* 11, 1700013 (2017).
99. Gellie P, Barbieri S, Lampin JF et al. Injection-locking of terahertz quantum cascade lasers up to 35GHz using RF amplitude modulation. *Opt Express* 18, 20799–20816 (2010).
100. Mottaghizadeh A, Gacemi D, Laffaille P et al. 5-ps-long terahertz pulses from an active-mode-locked quantum cascade laser. *Optica* 4, 168–171 (2017).
101. Forrer A, Franckié M, Stark D et al. Photon-driven broadband emission and frequency comb RF injection locking in THz quantum cascade lasers. *ACS Photonics* 7, 784–791 (2020).
102. Liao XY, Li ZP, Zhou K et al. Broadband terahertz quantum cascade laser dual-comb sources under off-resonant microwave injection. *Adv Photonics Res* 3, 2100361 (2022).
103. Liu GB, Ma XH, Zhou K et al. Farey tree locking of terahertz quantum cascade laser frequency combs. *Light Sci Appl* 14, 147 (2025).
104. Barbieri S, Ravaro M, Gellie P et al. Coherent sampling of active mode-locked terahertz quantum cascade lasers and frequency synthesis. *Nat Photonics* 5, 306–313 (2011).
105. Consolino L, Nafa M, Cappelli F et al. Fully phase-stabilized quantum cascade laser frequency comb. *Nat Commun* 10, 2938 (2019).
106. Li H, Yan M, Wan WJ et al. Graphene-coupled terahertz semiconductor lasers for enhanced passive frequency comb operation. *Adv Sci* 6, 1900460 (2019).
107. Riccardi E, Pistore V, Kang S et al. Short pulse generation from a graphene-coupled passively mode-locked terahertz laser. *Nat Photonics* 17, 607–614 (2023).
108. Piccardo M, Chevalier P, Schwarz B et al. Frequency-modulated combs obey a variational principle. *Phys Rev Lett* 122, 253901 (2019).
109. Opačak N, Schwarz B. Theory of frequency-modulated combs in lasers with spatial hole burning, dispersion, and Kerr nonlinearity. *Phys Rev Lett* 123, 243902 (2019).
110. Yang Y, Burghoff D, Hayton DJ et al. Terahertz multiheterodyne spectroscopy using laser frequency combs. *Optica* 3, 499–502 (2016).
111. Sterczewski LA, Westberg J, Yang Y et al. Terahertz spectroscopy of gas mixtures with dual quantum cascade laser frequency combs. *ACS Photonics* 7, 1082–1087 (2020).
112. Rösch M, Scalari G, Villares G et al. On-chip, self-detected terahertz dual-comb source. *Appl Phys Lett* 108, 171104 (2016).
113. Rösch M, Beck M, Süess MJ et al. Heterogeneous terahertz quantum cascade lasers exceeding 1.9 THz spectral bandwidth and featuring dual comb operation. *Nanophotonics* 7, 237–242 (2018).
114. Li ZP, Wan WJ, Zhou K et al. On-chip dual-comb source based on terahertz quantum cascade lasers under microwave double injection. *Phys Rev Appl* 12, 044068 (2019).
115. Senica U, Gloor S, Micheletti P et al. Broadband surface-emitting THz laser frequency combs with inverse-designed integrated reflectors. *APL Photonics* 8, 096101 (2023).
116. Digiorgio V, Senica U, Micheletti P et al. On-chip, inverse-designed

- active wavelength division multiplexer at THz frequencies. *Nat Commun* **16**, 7711 (2025).
117. Li H, Li ZP, Wan WJ et al. Toward compact and real-time terahertz dual-comb spectroscopy employing a self-detection scheme. *ACS Photonics* **7**, 49–56 (2020).
 118. Zhao YR, Li ZP, Zhou K et al. Active stabilization of terahertz semiconductor dual-comb laser sources employing a phase locking technique. *Laser Photonics Rev* **15**, 2000498 (2021).
 119. Zhao YR, Li ZP, Ma XH et al. Stabilized terahertz quantum cascade laser dual-comb sources with a hybrid locking. *ACS Photonics* **11**, 520–528 (2024).
 120. Li ZP, Ma XH, Zhou K et al. Terahertz semiconductor dual-comb sources with relative offset frequency cancellation. *Laser Photonics Rev* **17**, 2200418 (2023).
 121. Liu BB, Zhou K, Liu GB et al. Terahertz tri-comb laser sources. *Laser Photonics Rev* **19**, e01255 (2025).
 122. Wu JR, He F, Guo XG et al. Frequency noise in terahertz dual-comb spectroscopy using two repetition-frequency stabilized and synchronized femtosecond lasers. *Infrared Phys Technol* **147**, 105796 (2025).
 123. Shi DC, Kim B, Jang H et al. Photonic comb-rooted synthesis of ultra-stable terahertz frequencies. *Nat Commun* **14**, 790 (2023).
 124. Tetsumoto T, Ayano F, Yeo M et al. 300 GHz wave generation based on a Kerr microresonator frequency comb stabilized to a low noise microwave reference. *Opt Lett* **45**, 4377–4380 (2020).
 125. Shahili M, Addamane SJ, Kim AD et al. Continuous-wave GaAs/AlGaAs quantum cascade laser at 5.7 THz. *Nanophotonics* **13**, 1735–1743 (2024).
 126. Razeghi M, Lu QY. Room temperature terahertz and frequency combs based on intersubband quantum cascade laser diodes: history and future. *Photonics* **12**, 79 (2025).
 127. Di Gaspare A, Song C, Schiattarella C et al. Compact terahertz harmonic generation in the Reststrahlenband using a graphene-embedded metallic split ring resonator array. *Nat Commun* **15**, 2312 (2024).
 128. Sun SM, Wang BC, Liu KK et al. Integrated optical frequency division for microwave and mmWave generation. *Nature* **627**, 540–545 (2024).
 129. Zhang CH, Pelini J, Russo SD et al. Dual-comb photoacoustic and photothermal spectroscopy: a comprehensive review. *Photoacoustics* **47**, 100789 (2026).
 130. Wang Z, Nie QX, Sun HJ et al. Cavity-enhanced photoacoustic dual-comb spectroscopy. *Light Sci Appl* **13**, 11 (2024).
 131. Giovannetti V, Lloyd S, Maccone L. Advances in quantum metrology. *Nat Photonics* **5**, 222–229 (2011).
 132. Schnabel R. Squeezed states of light and their applications in laser interferometers. *Phys Rep* **684**, 1–51 (2017).
 133. Herman DI, Walsh M, Kreider MK et al. Squeezed dual-comb spectroscopy. *Science* **387**, 653–658 (2025).
 134. Wan ZR, Chen Y, Zhang XX et al. Quantum correlation-enhanced dual-comb spectroscopy. *Light Sci Appl* **14**, 257 (2025).
 135. Shi HW, Chen ZJ, Fraser SE et al. Entanglement-enhanced dual-comb spectroscopy. *npj Quantum Inf* **9**, 91 (2023).
 136. Hariri A, Liu S, Shi HW et al. Entangled dual-comb spectroscopy. *Phys Rev X* **15**, 041009 (2025).
 137. Gu CL, Zuo Z, Luo DP et al. Passive coherent dual-comb spectroscopy based on optical-optical modulation with free running lasers. *Photonix* **1**, 7 (2020).
 138. Shen XL, Yan M, Hao Q et al. Adaptive dual-comb spectroscopy with 1200-h continuous operation stability. *IEEE Photonics J* **10**, 1503309 (2018).
 139. Zhu ZB, Ni K, Zhou Q et al. Digital correction method for realizing a phase-stable dual-comb interferometer. *Opt Express* **26**, 16813–16823 (2018).
 140. Sterczewski LA, Westberg J, Wysocki G. Computational coherent averaging for free-running dual-comb spectroscopy. *Opt Express* **27**, 23875–23893 (2019).
 141. Yu HY, Ni K, Zhou Q et al. Digital error correction of dual-comb interferometer without external optical referencing information. *Opt Express* **27**, 29425–29438 (2019).
 142. Eber A, Gruber C, Schultze M et al. Streaming self-corrected dual-comb spectrometer. *Opt Express* **33**, 35314–35325 (2025).

Acknowledgements

This work was supported by the National Science Fund for Distinguished Young Scholars (No. 62325509), the National Natural Science Foundation of China (Nos. 62405089, 62235019, 62505344, 62531005, 62575299, 62275258, 62305364, 62435017, and T2550072), the Quantum Science and Technology-National Science and Technology Major Project (No. 2023ZD0301000), the Scientific Instrument and Equipment Development Project of the Chinese Academy of Sciences (No. PTYQ2026YZ0050), the Science and Technology Commission of Shanghai Municipality (No. 23ZR1474000), the CAS Project for Young Scientists in Basic Research (No. YSBR-069), and the Autonomous Deployment Project of the State Key Laboratory of Materials for Integrated Circuits (Nos. SKLJC-Z2025-B03, SKLJC-Z2026-B02, and SKLJC-Z2026-C01).

Author contributions

Z.W. conducted the literature survey, organized the manuscript structure, and wrote the original draft. B.L. and Y.X. contributed to literature collection, information analysis, and manuscript preparation. Y.C., X.Z., D.Z., and Y.D. assisted in literature collection and organization. M.L., M.Y., H.L., and H.Z. provided scientific guidance, supervised the project, and contributed to manuscript revision. All authors reviewed and approved the final manuscript.

Competing interests

The authors declare no competing financial interests.



Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. To view a copy of this license, visit <http://creativecommons.org/licenses/by/4.0/>

©The Author(s) 2026.

Published by Editorial Office of *Opto-Electronic Technology*, Institute of Optics and Electronics, Chinese Academy of Sciences.

