

EDITORIAL

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Research highlight: breaking the shot noise limit of mid-infrared dual-comb spectroscopy using intensity-difference squeezing

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Abstract

Correlated twin combs generated by seeded four-wave mixing enable quantum correlation-enhanced dual-comb spectroscopy. High signal-to-noise ratio and comb-line-resolvable spectra in the mid-infrared molecular fingerprinting region is achieved by bridging intensity-difference squeezing and dual-comb spectroscopy.

Spectroscopy is a fundamental tool for investigating the internal structure of matter in basic scientific research [1], and serves as a widely used non-contact detection technique in engineering applications [2]. As a cutting-edge spectroscopic technique, dual-comb spectroscopy (DCS) employs two optical frequency combs with a slight repetition-rate difference to generate multi-heterodyne interference, enabling the down conversion of spectral information from the optical domain to the radio-frequency domain. DCS combines high resolution, broad spectral coverage, thereby driving advances in both fundamental research and field-deployed applications [3–8].

Figure 1 summarizes key milestones in the development of DCS, focusing on its extension into the mid-infrared region and advancements in sensitivity. As an active detection technique, the spectral range of DCS is inherently constrained by the spectral bandwidth of the comb sources. Conventional DCS typically operated in the near-infrared region, while extending it into the mid-infrared—where many molecules show absorption

peaks—remains a substantial challenge for high-performance applications [9, 10].

The most direct approach for generating mid-infrared frequency comb sources is nonlinear conversions, such as difference frequency generation (DFG) [11–13] or optical parametric oscillation (OPO) [14, 15], which can shift the original near-infrared comb to the mid-infrared region. Moreover, compared to their well-developed near-infrared counterparts, mid-infrared photodetectors typically suffer from lower sensitivity, higher noise levels, and slower response times, which directly limit the signal-to-noise ratio (SNR) of mid-infrared DCS [16].

Upconversion detection technique [17], which converts the mid-infrared comb carrying gas absorption information to the near-infrared region via DFG, can partially mitigate this limitation. However, even in the well-developed near-infrared region, DCS—characterized by broadband and parallel detection—is inherently more susceptible to noise compared to tunable single-frequency lasers spectroscopy [18, 19]. Despite offering rapid single-shot acquisition, DCS typically relies on long-term averaging to achieve a satisfactory SNR [3, 20]. This bottleneck constrains dual-comb spectroscopy's applications with low optical power or trace samples.

Recently, considerable efforts have been made to push the sensitivity limits of DCS. Nathalie Picqué et al. [21, 22] obtained dual-comb interferograms under extremely low incident optical power using single-photon detectors. This approach relies on long-duration photon

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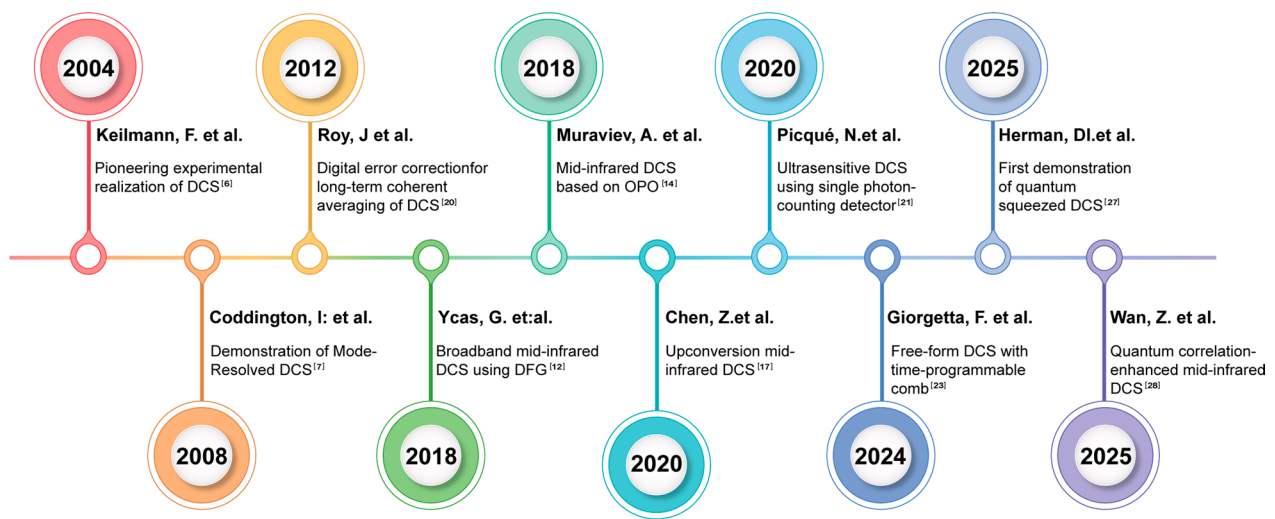


Fig. 1 Milestones in the development of dual-comb spectroscopy (DCS): extending into the mid-infrared region and pushing sensitivity limits

counting, which hinders its applicability in dynamically or rapidly changing scenarios. Fabrizio R. Giorgetta et al. [23, 24] employed two programmable combs to construct a free-form DCS system, mitigating the trade-off between spectral bandwidth and SNR. However, it is still fundamentally limited by the inherent shot noise of the coherent state combs.

Squeezing, as a representative quantum technique, provides promising avenues for surpassing sensitivity limits in classical interferometric system [25, 26]. Herman et al. [27] first demonstrated the metrological advantage of quantum mechanically squeezed comb interferometry. Enhancing the sensitivity of hydrogen sulfide molecular detection has been achieved based on squeezed dual-comb spectroscopy in the near-infrared. Subsequently, in a newly published paper in *Light: Science & Applications*, an up-conversion mid-infrared DCS scheme incorporating intensity-difference squeezing (IDS) has been proposed and demonstrated [28].

As illustrated in Fig. 2, Zhuoren Wan et al. proposed a quantum correlation-enhanced mid-infrared DCS configuration, combining the advantages of phase-insensitive IDS and up-conversion detection. To generate quantum-correlated twin combs, a near-infrared electro-optical comb is launched into a piece of highly nonlinear fiber alongside a synchronous pump comb with a certain wavelength difference. Due to the seeded four-wave

mixing, an amplified local comb and its accompanying idler comb are generated synchronously. In the detection stage, a weak-power mid-infrared signal comb carrying molecular fingerprint information is combined with the local comb to generate a conventional dual-comb interferogram. Meanwhile, the intensity noise signal of the correlated idler comb is detected and subtracted to cancel the quantum-correlated noise via IDS. In the experiment, the proposed quantum correlation-enhanced DCS achieves ~2-dB improvement in SNR beyond the shot-noise limit. Comb-line-resolved spectra with high resolution and high fidelity in the 3- μm mid-infrared molecular fingerprinting region have been achieved.

Recently, cutting-edge integrated nanophotonics technologies have accelerated the development of chip-scale DCS [29, 30]. Compared to conventional bulk dual-comb platforms, chip-based implementations often face greater limitations in terms of optical power and signal quality. Integrating dual-comb spectroscopy with quantum optics holds promise for overcoming these key technical bottlenecks. In the future, with the advancement of emerging artificial intelligence technologies [31, 32], it is expected to build a miniaturized and high-performance platform for broader dual-comb applications such as chemical analysis [33], imaging [34], and ranging [35].

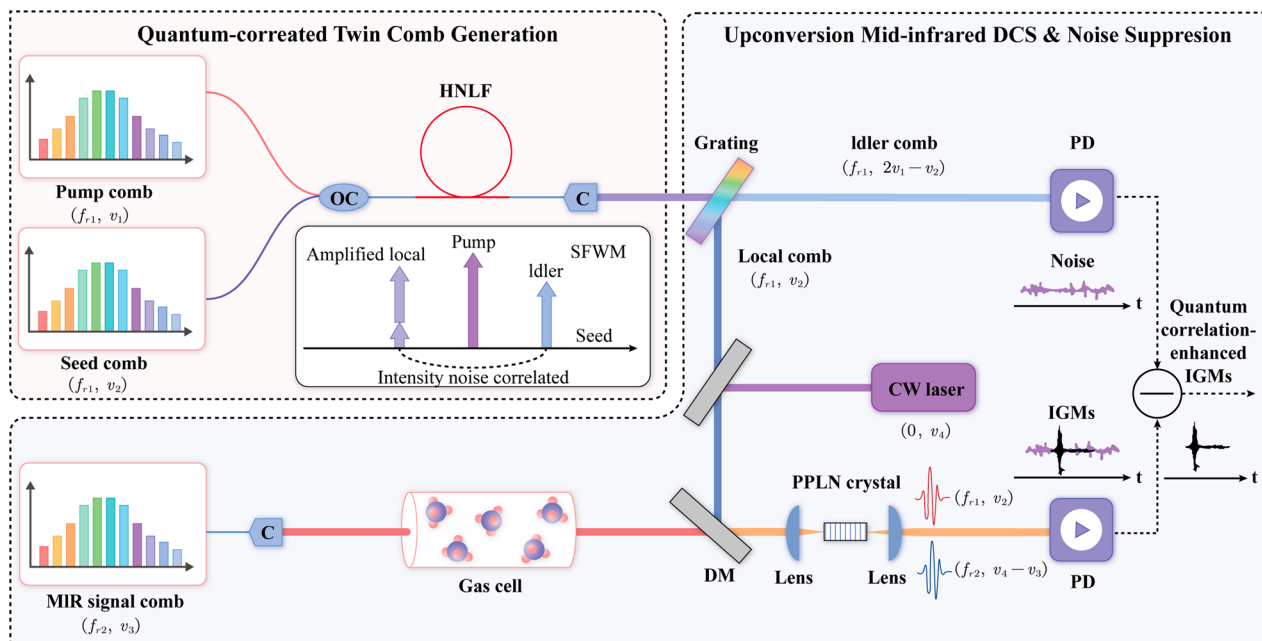


Fig. 2 Schematics of the quantum correlation-enhanced mid-infrared DCS using upconversion detection and intensity-difference squeezing (reproduced from [28]). OC: optical coupler. HNLF: highly nonlinear fiber. C: collimator. SFWM: seeded four-wave mixing. PD: photodetector. CW: continuous wave. IGMs: interferograms. PPLN: periodically poled lithium niobate. DM: dichroic mirror. f_{r1}, f_{r2} : repetition frequencies of dual combs. v_1, v_2, v_3, v_4 : carrier frequencies of pump comb, seed comb, signal comb and CW laser

Author contributions

GW and HY jointly wrote this editorial paper. All authors read and approved the final manuscript.

Data availability

The data supporting the figure and other findings in this paper are available from the corresponding author upon reasonable request.

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