TOPICAL REVIEW

Dual-comb generation from a single laser source: principles and spectroscopic applications towards mid-IR—A review

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Abstract

Dual-comb spectroscopy (DCS) is a promising approach for real time, high precision, and high sensitivity Fourier-transform spectroscopy. DCS requires a pair of tightly phase-locked optical frequency combs (OFCs). The intrinsic complexity hinders practical application of this technique. Recently, there is a new trend of generating a pair of OFCs with slightly different repetition rate from a single laser cavity, known as dual-comb lasers. Shared laser cavity cancels common-mode technical noises passively and mutual coherence between the OFCs is well maintained. Given that the cumbersome phase locking systems are avoided, dual-comb lasers provide an attractive alternative to a pair of independent OFCs. In this review paper, we summarize the latest laser cavity multiplexing schemes that allow dual-comb generation in solid state lasers, fiber lasers as well as in on-chip waveguide lasers. Their noise characteristics as well as the impact on the reachable performances in dual-comb applications, have been thoroughly discussed. Next, the recent progress of DCS based on single dual-comb lasers have been presented, unfolding the advantages of implementing ultrahigh precision molecular spectroscopic experiments with a simple setup. In particular, DCS towards mid-IR fingerprint region is emphasized. Finally, there is an outlook on the new trends of single dual-comb lasers working in mid-IR. The advances in DCS by combining multiple-comb mode-locking technique and recent emerging precision spectroscopic detection methods are also expected.

1. Introduction

Molecular absorption spectroscopy has been widely used in gas content determination, combustion diagnosis, human breath analysis, and so on. Particularly, as the ro-vibration absorptive features of most molecules lie in the mid-infrared (mid-IR) range, spectroscopy in the mid-IR attracts the most interest and significantly actuates the development of the conventional Michelson-based Fourier-transform infrared (FTIR) spectrometers [1]. When the thermal-optical source widely used in FTIR is replaced by a fully stabilized optical frequency comb (OFC), the performance could be greatly improved. Literally, an OFC [2, 3] is a phase-locked pulse train in the time domain, which corresponds to a broadband spectrum of evenly spaced coherent spectral lines with well-determined absolute frequencies in the frequency domain. Owing to the optimization of intrinsic noise performance and well-established phase locking technique, the repetition rate frequency $f_{\text{rep}}$ and carrier-envelope offset (CEO) frequency $f_{\text{CEO}}$ of an OFC could be well stabilized such that the comb linewidth is $< 1$ Hz [3–5]. High precision spectroscopy is enabled by the link from molecular absorption in optical frequency to the routinely measurable radio frequency (RF) via OFC [6].

However, even OFC-based FTIR is incapable of fulfilling a growing demand on spectroscopic measurement in terms of a fine spectral resolution at a high update rate, mainly limited by the scanning mechanical delay line introduced in one branch of the Michelson interferometer. Substituting of another

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OFC with slightly offset repetition rate for the mechanical delay line is one elegant approach to greatly improve the update rate and the resolution, which is recognized as dual-comb spectroscopy (DCS) [7–11].

The realization of dual-comb spectroscopy relies on asynchronous optical sampling (ASOPS) in the time domain, as shown in figure 1(a). Unlike the synchronous sampling which typically introduces different delay time with mechanical delay line, asynchronous optical sampling adopts two pulse trains with slightly different pulse repetition rates. As a result, ASOPS typically exhibits superior resolution/precision within a much shorter acquisition time. In dual-comb implementations, the update rate of measurement corresponds to the offset repetition rate. One frequency comb, with repetition rate of \( f_{\text{rep}} \), acts as the local oscillator (LO), while the other one with a repetition rate of \( f_{\text{rep}} + \Delta f_{\text{rep}} \) is the signal to be measured. When these two pulse trains are combined, a temporally varying delay between the two pulses is automatically generated, playing the role of mechanical scan in conventional Fourier transform spectrometers. The temporal sampling interval is determined by \( \Delta t_{\text{sampling}} = \Delta f_{\text{ref}} / (f_{\text{rep}} \cdot (f_{\text{rep}} + \Delta f_{\text{rep}})) \). After \( N = f_{\text{rep}} / \Delta f_{\text{rep}} \) LO pulses or \( N + 1 \) signal pulses, an equivalent sampling is completed. The required time duration for a single scan is calculated to be \( T = 1 / \Delta f_{\text{rep}} \). In other words, ASOPS effectively stretched the picosecond optical pulses by \( N \) times in time domain. Taking a laser with mega-Hertz repetition rate as an example, if the offset repetition rate between the LO and the signal \( \Delta f_{\text{rep}} \) is tuned around several kiloHertz, the amplification factor \( N \) is in the order of \( 10^5 \)–\( 10^6 \). Therefore, the generated signal in the order of nanoseconds can be directly detected by a photo-diode.

From the perspective of the frequency domain, two pulse trains with slightly different repetition rates can be expressed as two frequency combs in the optical frequency band with different frequency intervals, illustrated in figure 1(b). The combined pulses are mixed in the photo-diode, and the multi-heterodyne effect makes up another RF frequency comb between DC and \( f_{\text{rep}} / 2 \), limited by the Nyquist frequency, with an interval of \( \Delta f_{\text{rep}} \). The higher order difference frequencies make up the spectrum beyond \( f_{\text{rep}} / 2 \). With respect to the Nyquist frequency limit, the spectral bandwidth of the signal under test is limited by the
combination of repetition rate \(f_{\text{rep}}\) and the offset repetition rate \(\Delta f_{\text{rep}}\). When the optical spectrum is too wide, the down-converted radio frequency will exceed the boundary of DC to \(f_{\text{rep}}/2\), aliasing occurs. As the optical spectrum is connected with the radio spectrum by amplification factor \(N\), the optical spectrum should not exceed \(f_{\text{rep}}^2/(2\Delta f_{\text{rep}})\) in order to avoid aliasing.

DCS could be fulfilled with high update rate, fine resolution, and ultra-high precision, on condition that these two combs are mutual coherent, which is typically guaranteed by tightly phase-locking the two combs [8, 11–13]. Compared to the conventional spectrometers in dispersive configuration or FTIR, OFC based dual-comb spectroscopy excels in frequency resolution, accuracy, signal-to-noise ratio, and acquisition speed [10, 14]. Till now, DCS based on various phase locked laser sources, such as mode-locked lasers [5, 9, 15], inter-band cascade lasers [16], quantum cascade lasers [17–21], microresonators [22, 23] and EO-modulated combs [24] have been demonstrated.

Apart from the approaches by actively phase-locking two lasers, another trend is to excite a double-soliton operation state directly from a single mode-locked laser. Considering that the two pulses oscillate in a single cavity, mutual coherence is passively maintained, without the need of sophisticated and bulky phase-locking electrical sub-systems [25]. Slight offset repetition rate can be obtained via circulation-direction-multiplexing [26, 27], polarization-multiplexing [28, 29], wavelength-multiplexing [30, 31], cavity-space-multiplexing which relies on a compound laser cavity [32], as well as spatial mode multiplexing [33]. The greatly simplified implementation for DCS is referred as dual-comb laser technique. Until recently, dual-comb laser based spectroscopic application has been widely validated in the near-IR spectral region [26, 27, 30], while the most promising band is located beyond 2 \(\mu\)m where a number of important ‘finger-print’ of molecules exists [34]. To this end, dual-comb lasers working at mid-IR is necessary.

In this review, we describe the generations and spectroscopic applications based on a single dual-comb laser aiming at mid-IR spectral region. The review is organized as follows. Section 2 provides a brief introduction to fundamental approaches to generate dual-comb in one laser cavity among solid-state lasers, fiber lasers, microresonators, etc. Section 3 discusses noise characteristics of the two optical solitons circulating in dual-comb lasers in terms of relative timing jitter, comb-line broadening and CEO frequency noise. These noises could make non-negligible contribution to asynchronous optical sampling process and the related DCS applications. Section 4 provides iconic DCS demonstrations toward the mid-IR region. Recent DCS experiments based on single free running dual-comb lasers are emphasized. Finally, in section 5, a selection of near-future trends and prospects of mid-IR dual-comb laser sources as well as their DCS applications are envisioned.

### 2. Generation

#### 2.1. Principle

Various routines have been verified to generate dual-combs with minor repetition rate difference from a single mode-locked laser cavity. Among them, wavelength-multiplexing, polarization-multiplexing, circulation-direction-multiplexing and cavity-space-multiplexing are the most representative techniques discussed in literatures. In wavelength-multiplexing technique, two pulses with distinct central wavelengths are oscillating in one laser cavity simultaneously, as shown figure 2(a). Cavity group velocity dispersion (GVD) makes them oscillate in the laser cavity asynchronously with slightly different repetition rate. To achieve dual-wavelength operation from a single mode-locked laser, one could either implement bandpass spectral filters (like fiber Bragg gratings), or equivalent spectral filtering elements (e.g. Lyot filters, Sagnac filter, intracavity tapper fiber, etc) into the cavity. In polarization-multiplexing technique, two pulse trains with orthogonal polarization states coexist in one laser cavity. Through introducing a birefringent medium e.g. a piece of polarization-maintaining (PM) fiber into the cavity, the refractive index difference between ordinary and extraordinary axis (fast axis and slow axis for PM fiber) could result in offset repetition rate between pulse trains \(a\) and \(b\), as shown in figure 2(b). Besides, the circulating directions in a propagating wave cavity can also be multiplexed with clockwise and counter-clockwise operation, as shown in figure 2(c).

In this case, the difference in repetition rates between the two pulse trains either links to direction-dependent Kerr non-linearity in solid state lasers [26], or originates from distinct central wavelength of CW an CCW laser pulses introduced by direction-dependent absorption in gain fiber, non-linear phase shift, etc in fiber lasers [27]. This is termed as circulation-direction-multiplexing. Apart from those having a strict single cavity, some dual-comb schemes rely on a compounded cavity, which are termed as cavity-space-multiplexing, as shown in figure 2(d).
2.2. Dual-comb in solid-state mode-locked lasers

During the last 40 years, there have been tremendous progress on the solid-state mode-locked lasers owing to the advances in doped laser crystals and mode-locking mechanisms. Among them, researches on Ti:sapphire mode-locked lasers keep expanding until today due to its superior performance in the few-cycle pulse generation directly from an oscillator, the broadband gain spectrum, and the extremely low optical phase noise [35].

The first dual-comb generation in solid-state mode-locked laser was reported in 1993 from Ti:sapphire lasers by AT&T Bell laboratories [36]. It adopted a wavelength-multiplexing scheme and the intracavity band gap filter was functioned by selecting two parts of spatially dispersed optical spectrum with a slit inside the laser cavity. Both Ti:sapphire [37, 38] and Nd:CNGG lasers [39] demonstrated such a dual-color operation. Two-color pulse trains with central wavelength separations of 60 to 80 nm were realized. Dual-color mode-lockings in Nd:LNGG laser with SESAM were also reported in reference [40–42]. Note that early studies did not noticed the importance of differential repetition rate for dual-comb applications. They developed the dual-color laser sources mainly for timing synchronization. To this end, the above works usually used a compounded laser cavity. Therefore, the repetition rate difference due to GVD can be zeroed by tuning the length of the non-common cavity arms separately.

Until 2016, T Ideguchi et al reported a Ti:sapphire mode-locked bidirectional ring femtosecond laser, which generated two broadband OFCs with slightly different repetition rate without birefringent crystal [26]. ∆frep between two pulses could be tuned from 100 to 800 Hz via the displacement of Ti:sapphire crystal, cavity mirror or focusing lens of pump. On the other hand, dual-comb generated from semiconductor thin disk mode-locked laser was first reported by S M Link et al [28, 43, 44]. Through an intracavity birefringence crystal, two orthogonal-polarized pulses with similar spectra centered at 968 nm was produced with a ∆frep of 4 MHz. Repetition rate difference at MHz level is capable of shortening the acquisition time to 2.5 µs in time-dependent interferogram detection. Meanwhile, as the comb spacing is fairly large, the energy per comb-line is higher than that of typical fiber lasers, resulting in superior signal-to-noise ratio even for single-shot interferogram measurement.

Apart from the generation of dual-comb directly from a mode-locked laser, optical parametric oscillator (OPO) pumped by two individual combs also showed potential to generate a dual-comb with slightly different repetition rates [45]. Sharing one common OPO cavity for frequency-conversion of two free-running seeded lasers could improve the dual-comb spectroscopy measurement performance by avoiding the random drifts in two independent OPO cavities [46, 47].
2.3. Dual-comb in fiber mode-locked lasers

Fiber-based mode-locked lasers, especially erbium-doped (Er) fiber mode-locked lasers at telecommunication band, become more feasible sources for further practical applications owing to their low-cost pump diodes, environmental robustness, and compactness. Moreover, versatile mode-locking mechanism and pulse shaping dynamics inside fiber laser enable ample approaches towards dual combs or even triple combs generation from one single laser [30, 48, 49].

2.3.1. Wavelength-multiplexing

Wavelength-multiplexing is the most direct way to expand the available optical channels in fiber lasers. The introduction of intracavity filter could lead to the dual-wavelength mode-locking in one fiber laser with asynchronous pulse trains. As illustrated in figure 3, the dual-peak net gain spectrum with the balance of gain, loss, and filtering effect plays crucial roles in the formation of dual-color pulses. Benefiting from the central wavelength difference of two pulse trains, their group velocities show slight difference because of the optical fiber’s GVD. Accordingly, the offset repetition rate $\Delta f_{\text{rep}}$ is determined by the following expression:

$$\Delta f_{\text{rep}} = \frac{1}{D \cdot L \cdot \Delta \lambda},$$

where $D$ is the dispersion parameter in units of ps/(nm $\cdot$ m), $L$ is the optical-path length of the cavity, and $\Delta \lambda$ is the gap of center wavelength in between.

One of the practical intracavity filter is Lyot filter. Fiber based Lyot filter, which consists of polarizers, PM fibers and polarization controllers, is an optical device with wavelength-dependent transmission. Generally speaking, Lyot filter is a kind of interferometric filter. Utilizing the birefringence of single-mode fiber or directly splicing a piece of PM fiber into the laser cavity, implementation of fiber Lyot filter in non-linear polarization evolution (NPE) scheme for dual-wavelength mode-locking have been widely adopted in Er: fiber lasers [50–62], Yb: fiber lasers [63, 64] and Bi: fiber lasers [65]. Note that in a Lyot filter, two pulses propagate in the same direction. On the other side, in a Sagnac filter, two pulses in a Sagnac loop propagate in opposite directions and finally interfere at the output, also functioning as a wavelength-dependent optical filter. Dual-comb generation using intracavity fiber Sagnac filter was demonstrated in reference [31, 66, 67]. Besides these interferometric fiber filters [68], cavity loss tuning [69], intracavity tapper fiber [70] and intracavity mode select coupler [71] could also function as equivalent spectral filters to realize dual-comb operation in fiber mode-locked lasers.

2.3.2. Polarization-multiplexing

In single-mode fibers, there are two orthogonally polarized modes co-existing with the same spatial distribution. For an ideal fiber these two modes are degenerated because they share an identical effective refractive index, meaning $n_x = n_y$, where $n_x$ and $n_y$ are the modal refractive indices for the two orthogonally polarized states [72]. In reality, all fibers exhibit unintentional variations in the fiber core shape and asymmetric stresses along the fiber length. As a result, modal birefringence emerges for $n_x \neq n_y$ [73]. Vector soliton, which describes soliton pair in birefringent fibers on the two polarization axes, has been intensively studied since the first discovery [74–77]. The two orthogonal components in a vector soliton share a same non-linearity induced group velocity. However, if the mismatch of the velocities between the solitons becomes in-compensable, dual-comb with an offset repetition rate forms. Typically, it is not easy to get the two polarization components propagating with two different group velocities in fibers. Because in single-mode fiber, the fast and slow axes are not well defined, while in polarization-maintaining (PM) fiber, the cavity length $L$ is far beyond the fiber beat length $L_B$, which is related to the polarization mode dispersion in fibers. The beat length $L_B$ can be calculated through $L_B = \frac{\lambda}{|n_x - n_y|}$. As a result, researchers managed to
obtain a dual-polarization dual-comb laser by inserting a short length of PM fiber into a cavity mostly made of single-mode fiber [29, 78–80]. The offset repetition rate, typically in the range of several hundreds of Hertz, is determined by the cavity length and a dimensionless parameter used to measure the modal birefringence in fibers $B_{\text{mod}} = |n_x - n_y|$. In contrast to the most reported dual-wavelength dual-comb lasers, dual-comb lasers with dual-polarization can output pulses with overlapping spectrum thus enabling intrinsic spectral coherence.

2.3.3. Circulation-direction-multiplexing
Bi-directional operation of fiber laser has been realized since 2008 [81]. Without the restriction of an optical isolator inside a propagating-wave laser cavity, clockwise and counter-clockwise light fields can oscillate simultaneously. In reference [81], the bi-directional fields are either circulating with slightly different propagating velocities, or with the same group velocity but different carrier frequencies. Strictly, bi-directional dual-comb lasers do not explicitly decide which physical quantity of the pulse are multiplexed. The non-symmetrical gain distribution is commonly regarded to contribute to the generation of offset repetition rate, which also reflects on the difference of spectra most of the time [27, 48, 81–86]. However, compared to the dual-color dual-comb lasers, the difference of spectra between the two combs are minor and no spectral gap in between exhibits. As a result, no explicit solution can be derived for the roundtrip time difference of the two combs. Typically, the offset repetition rate is smaller compared to the aforementioned two kinds of dual-comb lasers.

2.3.4. Cavity-space-multiplexing
Although common optical-paths are guaranteed in the above-mentioned schemes, the implementation as well as the tuning range is not satisfactory. To introduce sufficient flexibility into the dual-comb laser, efforts are paid to laser designs with a segment of non-common optical path, as reported in [32, 87–93]. To this end, the offset repetition rate can be tuned in a relatively larger range, suitable to extensive applications. To reduce the strong interaction between the two combs, two individual saturable absorbers are adopted to separate the pulse generation process [32, 92, 94]. In reference [93], two segments of gain fibers are used to isolate the pulse dynamics between the two pulse trains. Actually, the dual-comb laser can output polarization-multiplexed pulses with either distinctive or similar spectra, which possesses potential in various applications.

2.3.5. Extra-cavity fiber delay methods
In 2018, Carlson et al presented an extra-cavity fiber-delay method to realize asynchronous dual-comb using one femtosecond laser [95]. In this case, the femtosecond laser’s repetition rate is alternated at $f_{\text{rep}}$ or $f'_{\text{rep}}$ with a periodical rate of $f_{\text{mod}}$. The output of the comb is sent into an asymmetric fiber Michelson interferometer with a fixed length of optical fiber in one arm. In this way, the output pulse train heterodynes with its own delayed one, resulting in two pulse trains with determinate $\Delta f_{\text{rep}}$, which is tunable depending on the modulation depth of the cavity length. Modulation frequency $f_{\text{mod}}$ needs to be carefully chosen to match the length of the fiber delay to produce the dual-comb interferograms. Distance measurement and spectroscopy were also demonstrated using this dual-comb system.

2.4. On-chip techniques
Dual-comb integration in a single chip makes it suitable for future lab-on-a-chip applications. Asynchronous pulse trains in Er-doped ZBLAN glass mode-locked lasers was first realized through sharing a same laser cavity in 2017 [96]. In this setup, $\Delta f_{\text{rep}}$ is determined by the slight optical path difference that two pulse trains are experiencing. Meanwhile, dual-comb operation in a single microresonator has also been reported owing to the recent advances in chip-based non-linear photonics. Dual-comb generation in microresonators mainly relies on counter-propagating dissipative optical solitons inside high-Q microresonator pumped by bidirectional continuous-wave lasers [97]. Alternatively, dual-comb generation is realized by polarization-multiplexing via exploiting both transverse electric (TE) mode and transverse magnetic (TM) mode in one microresonators [98]. For applications requiring more than two combs, the multiplexing could even be realized by the excitation of multi-spatial-modes. E Lucas et al show the scalability of mode-multiplexing by simultaneously pumping three different mode families [33]. In the demonstration, two modulated sidebands of the continuous wave are launched in the clockwise direction while the original continuous wave is launched in the counter-clockwise direction. Therefore, three soliton combs in three different spatial modes are simultaneously excited in a MgF$_2$ microresonator. Benefiting from extremely short optical cavity, the repetition rates of microresonators are always at $\sim 10$ GHz level with at least kHz level offset repetition rate.

Figure 4 summarizes the recent advances of dual-comb mode-locking in solid state, fiber, and microresonator laser sources by various multiplexing approaches. Most demonstrations are within the recent
few years, showing the prosperity in this research field. The mode-spacing and offset repetition rate of each laser can be read from this figure. The obtainable mode-spacing based on dual-comb mode-locking technology extends from several MHz to tens of GHz, while the available offset repetition rate ranges from several Hz to hundreds of MHz, satisfying various special demands in dual-comb molecular spectroscopic applications.

3. Stability of dual-comb lasers

Common-mode noise suppression based on a shared cavity is regarded as the major advantage of dual-comb mode-locked lasers. There are two degrees of freedom in an optical frequency comb, namely, the repetition rate $f_{\text{rep}}$ and carrier envelope offset frequency $f_{\text{CEO}}$. It has been testified by many literatures that even though individual $f_{\text{rep}}$ ($f_{\text{CEO}}$) of a free-running dual-comb mode-locked laser drifts over time, the offset between the two combs $\Delta f_{\text{rep}}$ ($\Delta f_{\text{CEO}}$) shows superior long-term stability [26, 30, 32]. For example, figure 5 (a) and (b) are $f_{\text{rep}}$ and $\Delta f_{\text{rep}}$ fluctuations in a wavelength-multiplexed dual comb thulium:fiber mode-locked lasers, respectively [31]. Though each $f_{\text{rep}}$ drifts over tens of Hertz during 600 s owing to environmental perturbations, $\Delta f_{\text{rep}}$ remains relatively stable with a standard deviation of 0.11 Hz. Similar behavior of the temporal fluctuations of $f_{\text{CEO}}$ and $\Delta f_{\text{CEO}}$ has been verified in reference [32], measured in self-referencing scheme. The passively stabilized $\Delta f_{\text{rep}}$ and $\Delta f_{\text{CEO}}$ reflect intrinsic mutual coherence between the two pulse trains, which is essential for dual-comb applications.

Despite of the outstanding long-term stability, some kinds of de-phasing effects were indeed observed in dual-comb mode-locked laser sources. On the one hand, periodic colliding between the two pulse trains inside the cavity inevitably destabilizes mode-locking. On the other hand, we find that ASE noise, which is quantum noise in nature, cannot be eliminated by sharing a single cavity, and set a fundamental limit on the reachable performance in time- and frequency-domain dual-comb applications. Details of these effects will be discussed in the following.

3.1. Pulse collision inside the laser cavity

In dual-comb lasers, as two pulse trains are traveling simultaneously intracavity with a slightly different propagation velocity, these two pulse trains will collide with each other every several kilo-roundtrips when they are co-propagating inside the laser cavity, or every roundtrip when they are counter-propagating. As a result, although the intracavity pulse dynamics are independent when two pulses are far apart, situation may be more complicated when two pulses are close enough. On the other hand, despite the obvious advantages of common-mode noise cancellation and simple implementation, the two pulse trains oscillating in the same laser cavity indeed interact with each other and may have an impact on high precision dual-comb applications. The study of the underlying pulse interaction mechanisms is within the scope of multi-soliton solutions of the coupled Ginzburg-Landau equation (CGLE) [99]. Intensive attention has been paid on the
formation and dynamic evolution of multi-solitons [100, 101]. Transient soliton collision in a passively
mode-locked fiber laser has also been investigated and regarded similar to the Feynman diagram [101].
However, most of these works discussed the solitons propagation with the same group velocity in the stable
state. The first experimental observation of periodic pulse collision in a dual-comb laser was reported in a
stretched-pulse fiber laser by [102], where consecutive collisions took place between a single pulse and a
two-pulse bound state, addressing that the bound states can remain unaltered after the collision. Later
through theoretical analysis, they found that in these nonconservative systems, the pulses do not fully overlap
in the course of a collision, quite different from those in integrable systems. The exchange of a bond takes
place by interacting only through their tails, which is termed ‘anti-crossing collision’ [103]. Almost at the
same time, ‘elastic’ collision was observed between a dissipative soliton pair and a soliton singlet by [104],
which again obeys the rules of the anti-crossing collision. To explore the evolution and interaction of
dual-comb solitons, the formation of dual-wavelength soliton has been numerically and experimentally
investigated by [105]. Recently, researchers managed to visualize the collision-induced soliton self-reshaping
process between dual-color solitons, through the technique of time-stretched dispersion Fourier transform
(TS-DFT) and numerical simulation [58]. Consider the different multiplex method in dual-comb lasers,
richer soliton interaction mechanisms are expected in those specific lasers [106, 107].

Intracavity pulse interaction indeed leads to instability of dual-comb mode-locked lasers, as revealed in a
recent $f_{\text{CEO}}$ measurement experiment [32]. A bi-directional Er-fiber mode-locked laser has been used. The
stability of two $f_{\text{CEO}}$s has been characterized by a phase noise analyzer. The result shows that additional noise
peaks around the offset repetition rate are visible in dual-comb operation state, indicating that dephasing
interaction occurred between the two combs inside the laser cavity.

3.2. Impact of quantum noise on dual-comb mode-locked lasers

3.2.1. Quantum-limited timing jitter

Timing jitter of optical pulses is defined as the short-term temporal deviation of pulse’s envelope from
perfectly periodic temporal positions. In free-running passively mode-locked lasers, the pulse timing
undergoes a random walk characteristic. Based on soliton perturbation theory, intracavity amplified
spontaneous emission (ASE) induced timing jitter for soliton pulses is obtained as [108, 109]:

$$ S_{\text{ASE,soliton}}^{\Delta f} = \frac{D_T}{(2\pi f)^2} \left( \frac{4D_T f_{\text{rep}} D_{\omega c}}{(2\pi f)^4 [ (2\pi f)^2 + \tau_{\omega c}^2] } \right) $$

where $f$ is Fourier frequency; $D_T$ is diffusion constants of ASE-induced group delay velocity; $D_{\omega c}$ is diffusion
constants of center frequency variations for optical pulses; $D$ is half of net cavity dispersion; $\tau_{\omega c}$ represents
the decay time for frequency perturbation; $f_{\text{rep}}$ is repetition of pulse train. The first term represents the
directly ASE-coupled timing jitter. The second term corresponds to timing jitter coupled by central
frequency fluctuation through cavity dispersion, well-known as Gordon-Haus jitter. Relative timing jitter
between the two pulse trains from the dual-comb mode-locked laser will lead to short-term fluctuation of
$\Delta f_{\text{rep}}$, thus degrade mutual coherence between the two pulse trains. Note that in a dual-comb laser, ASE
induced relative timing jitter which is quantum noise in nature, is uncorrelated, thus cannot be removed by
sharing a laser cavity. Particularly, in dual-wavelength lasers, the cavity net dispersion needs to be shifted
from zero so as to generate asynchronous pulses. The non-zero $D$ inevitably increases the relative timing
jitter originated from Gorden-Haus jitter.
Figure 6. (a) Principle of timing jitter estimation using ASOPS scheme. (b) Experimental results of timing jitter estimation. Inset shows the histogram of relative timing jitter in the stretched time scale when $T_p$ is set as $\sim 11 \mu$s. Reprinted with permission from reference [25], IEEE.

The ASE-limited timing jitter in passively mode-locked lasers is very small, typically $< 1$ fs. However, we have successfully characterized the relative timing jitter between the two optical solitons in a dual-wavelength fiber laser by using the inherent ASOPS process in the dual-comb laser [25, 110], as shown in figure 6(a). When the output pulse trains from the dual-wavelength dual-comb fiber laser are separated, one pulse train serves as local oscillator (LO) while the other pulse train is laser under test (LUT). The output from the LUT is interleaved by an asymmetric Michelson interferometer with one moving end mirror. In this way, $t_p$, which corresponds to the pulse-to-pulse interval in each interleaved pulse pair, could be adjusted. The standard ASOPS process is capable of effectively magnifying the laboratory time scale by a factor of $N = f_{\text{rep}} / \Delta f_{\text{rep}}$, mapping the time interval $t_p$ to $T_p$ in the stretched time scale, where relative timing jitter between LUT and LO will introduce uncertainty on $T_p$, which could be defined as ‘visual period jitter’ with $\sigma_{\text{visual}} = N \sigma_M$, where $\sigma_M = \sigma_0 \sqrt{M}$, $M \approx T_p / t_r$. Figure 6(b) shows that $\sigma_{\text{visual}}$ scales linearly with $T_p$.

The uncertainty on $T_p$ can be easily determined by standard electronics, and the period jitter $\sigma_0$ for the mode-locked laser can thus be retrieved by using $\sigma_0 = \sigma_{\text{visual}} / (N \cdot \sqrt{M})$. In the dual-wavelength fiber laser in reference [25], the obtained period jitter STD was $\sigma_0 = 0.82$ fs, which is typical for a quantum-limited soliton fiber laser, as shown in figure 6(b). Inset in figure 6(b) shows the histogram of relative timing jitter in the stretched time scale when $T_p$ is set as $\sim 11 \mu$s. The Gaussian distributed histogram with $\sigma_{\text{visual}} = 0.454$ ns confirms that, despite the inherent long-term stability is ensured by sharing one laser cavity, the two pulse trains inevitably suffer uncorrelated ASE noise, in the short timescale.

3.2.2. Quantum-limited relative comb linewidth

Fixed-point theory [111, 112] of mode-locked lasers depicts the mechanism of comb line broadening induced by intracavity ASE. Frequency noise power spectral density of comb line caused by ASE noise follows

$$S_{\nu, n}^{\text{ASE}}(f) = S_{\nu}^{\text{ST}} + (\nu_n - \nu_c)^2 (2\pi f)^2 S_{\Delta \nu}^{\text{ASE}}(f)$$

where $f$ is Fourier frequency, $\nu_n$ is the optical frequency of the $n$th comb line; $\nu_c$ is the center frequency of the OFC, $S_{\nu, n}^{\text{ASE}}(f)$ is the power spectral density of ASE-limited timing jitter. The first term corresponds to Schawlow-Townes limit originated from optical-carrier phase jitter. In most cases, Schawlow-Townes limit linewidth is extremely narrow and negligible compared with technical noise such as pump fluctuations induced comb line noise. The second term represents the comb line broadening contributed from ASE-coupled timing jitter.

Due to the uncorrelated feature of ASE-induced relative timing jitter between two pulses trains, the diffusion factor inevitably causes relative linewidth broadening in the dual-comb mode-locked laser. Given that the 3 dB linewidth of the $n$th comb mode can be estimated as $\Delta \nu = \pi S_{\nu, n}(f)$, the timing jitter induced...
Table 1. A comparison for performance of representative fully-stabilized dual-comb systems and dual-wavelength mode-locked fiber lasers.

| category       | reference | $\lambda_c$ [nm] | $f_{rep}$ [MHz] | $\Delta f_{rep}$ [kHz] | $\Delta f_{opt}$ [THz] | $\Delta \nu$ [Hz] |
|----------------|-----------|------------------|-----------------|-------------------------|-------------------------|-----------------|
| two lasers     | [12]      | 1550             | 100             | 1.10                    | 186–200                 | 100 kHz $^a$    |
|                | [114]     | 1960             | 115.23          | 0.069                   | 146–162                 | >3 kHz          |
|                | [115]     | 3260             | 100             | 0.13                    | 88–96                   | 5.3 MHz $^a$    |
| single laser   | [30]      | 1535             | 52.74           | 1.25                    | 194–196                 | 250 Hz          |
|                | [31]      | 1940             | 71.88           | 3.27                    | 154–155                 | 5.6 kHz $^a$    |

$^a$ Limited by acquisition time.

comb linewidth will increase quadratically apart from the comb center, according to equation (2). Recently, the elastic tape behavior of a bi-directional dual-comb ring laser was investigated by Lomsaze et al based on fixed-point theory [113]. In 2018, we showed that in a typical dual-wavelength dual-comb fiber mode-locked laser, the relative comb line broadening through quantum-limited timing jitter is estimated to be 650 Hz when the comb tooth is 12 nm (1.5 THz) away from the comb center [25]. This line broadening process will set an ultimate spectral resolution for dual-comb spectroscopic applications, as investigated based on a recent numerical simulation [25]. This effect is ignorable in most gas absorption spectroscopic experiments where Doppler-broadening is dominated. However, in the case of Doppler-free spectroscopy, the impact of quantum-limited line broadening should be definitely taken into consideration.

A comparison for representative dual-comb systems generated from two fully-stabilized mode-locked lasers and from a single dual-wavelength mode-locked fiber laser is shown in table 1, where $\lambda_c$ stands for the central wavelength, $\Delta f_{rep}$ the reported optical frequency coverage, and $\Delta \nu$ the relative linewidth between two combs. In two fully-stabilized mode-locked lasers [12, 114, 115], relative optical linewidth is routinely decided by the reference continuous-wave laser (typically at kHz level linewidth and possibly down to mHz by Pound–Drever–Hall (PDH) technique) as well as the performance of phase locked loop. While, in the case of dual-wavelength from one mode-locked fiber lasers [30, 31], relative linewidth between the two OFCs oscillating in the same cavity is above hundreds of Hertz, limited by non-common mode noise sources in the lasers as well as extra-cavity spectral broadening processes.

Considering the noise performance of individual down-converted comb lines in RF domain, the noise generated during optical-to-electrical conversion process would add excess noise to every comb lines [116]. As investigated in reference [117], shot-noise, thermal-noise, flicker noise and non-linearity generated by photo-detection devices could all deteriorate signal-to-noise ratio (SNR) of RF comb lines. In dual-comb spectroscopic applications, the limiting SNR is linked to dark current, Johnson noise of photo-detector as well as detector dynamic range [118]. Coherent averaging is the simplest yet most practical approach to remove Gaussian white detector noise to improve detection SNR and measurement resolution [9], while parallel detection over the full optical spectrum is beneficial for reducing the dynamic range limitation [118].

4. Dual-comb spectroscopic application towards mid-IR

Dual-comb spectroscopic applications have shown their great potentials in mid-infrared region, because the rotational and vibrational induced absorption peaks of most molecules are located in regions above 2 $\mu$m. In this region, non-linear optical frequency conversion provides the most flexible frequency coverage. To this end, thulium laser pumped orientation-patterned GaAs (OP-GaAs) optical parametric oscillator (OPO) producing 3–6 $\mu$m mid-IR laser has been adopted for dual-comb spectroscopy [119]. D T Reid et al successfully used the idler pulses from an OP-GaP OPO for absorptive spectrum detection of methane around 7 $\mu$m [120], and N R Newbury et al used difference frequency generation (DFG) in PPLN covering a spectrum from 2.6 to 5.2 $\mu$m for measuring a gas mixture of methane, ethane, acetylene, and nitrogen [121]. However, it is also regarded that in the mid-infrared region, mutual coherence between two laser systems built on non-linear frequency conversion is more challenging to achieve and to maintain [24].

The rapid developing on-chip nanophotonics also show great potential for mid-IR dual-comb spectroscopy. There are two approaches to extending the wavelength of on-chip nanophotonics into mid-IR. One approach is the conventional supercontinuum generation process pumped by near-infrared ultrafast pulses, not in fibers or bulk crystals, but in waveguides such as Si$_3$N$_4$ [122, 123]. In reference [123], Si$_3$N$_4$ waveguides, pumped at 1550 nm, could provide spectrally smooth, broadband OFC for gas spectroscopy in 2–2.5 $\mu$m range, which is known as the important atmospheric water window. Dual-comb spectroscopy was also conducted for the detection of CO and CO$_2$. The other approach is directly frequency combs generation from microresonator pumped by continuous wave in mid-IR, like reference [22]. A continuous wave OPO
emitting at 3 \( \mu \text{m} \) simultaneously pumped two microresonators with slightly different comb-line spacing. The resultant spectrum covered 2.6–4.1 \( \mu \text{m} \) with a comb-line spacing around 127 GHz. Benefited from the ultra-short cavity length, frequency comb generated from microresonator typically exhibits large comb spacing and relatively higher comb energy, which typically leads to higher SNR and faster acquisition in dual-comb detection.

Electro-optic modulation (EOM) of a continuous-wave laser is another routine approach to producing an optical frequency comb. In order to prepare a dual comb, a straightforward method is to beam split a single continuous-wave laser and modulate the two branches of lasers by two different RF frequencies with separate electro-optical modulators. However, constrained by the available laser source, only limited results have been reported in the mid-IR region. In 2017, a 3 \( \mu \text{m} \) idler was generated in DFG process with signal from 1550 nm dual-comb EOM laser source, where the absorption of methane and ethylene were measured at Doppler-limited resolution [24]. Electro-optic dual-comb spectrometer in the thulium amplification band is also reported, the spectrometer can resolve the absorption lines by carbon dioxide around 2 \( \mu \text{m} \) [124].

The above mentioned dual-comb spectroscopy experiments prove to be a powerful tool in mid-IR precision spectroscopy, except that two separate optical frequency combs are required. Recently, there has been a rapid increase in dual-comb spectroscopic applications based on various free-running dual-comb lasers. In 2016, dual-comb laser was first applied for spectroscopic measurement, where bi-directional Ti:sapphire, bi-directional Er-doped fiber laser, and dual-wavelength Er-doped fiber laser have been used as laser sources [26, 27, 30]. Particularly, in reference [30], picometer-resolution dual-comb spectroscopy with a single free-running Er-fiber laser has been demonstrated at 1550 nm wavelength, benefited from the superior stability of the dual-comb fiber laser against common-mode environmental noise.

To match the absorption band of methane, J Genest et al further broadened the output wavelength of a dual-comb Er-doped laser to 1670 nm [125]. The laser source is a space-multiplexed free-running chip-based dual-comb laser, producing an optical comb spacing of 968 MHz and an offset repetition rate of 27 kHz. The dual-comb laser was used to interrogate the complex transmission spectrum a methane-filled gas cell. A 1.28 s sequence of interferograms was measured and phase-corrected using a self-sufficient correction algorithm seeded only by the interferograms. The associated transmission spectrum was compared to HITRAN, yielding residuals only limited by photo-detector non-linearity.

To further extend the operation wavelength toward mid-IR region, our group conducted a proof-of-principle experiment with a single thulium-doped dual-comb fiber laser [31, 126]. The implementation of the dual-comb laser was based on wavelength-multiplexed scheme, as illustrated in figure 7(a). With a comb spacing of 72 MHz and an offset repetition rate around 3 kHz, the sampling process provided a magnification factor of 24 k, and an acquisition duration of single frame interferogram around 0.3 ms. With temporal averaging of data in 1 s, the absorption lines of water vapor around 1940 nm were resolved. The SNR is greatly enhanced in the coherent averaging process, reflecting the intrinsic stability of the two combs. Comb-line resolvability was also checked in the experiment by taking advantage of the intrinsic mutual coherence between the two combs. The fine structure of the comb-lines is clearly resolved in figure 7(b). Details can be reached in reference[31], and the comb linewidth is identical to the reverse of sampling duration, which indicates that the obtainable spectral resolution is still limited by the temporal sampling duration.

5. Outlook

In the future, there are two trends on spectroscopic applications based on single dual-comb laser sources. Firstly, in order to meet the practical applications of Mid-IR molecular spectroscopy, the increase of mid-IR dual-comb laser sources is expected. Secondly, the performance of dual comb spectroscopy will be significantly advanced by combining dual-comb mode-locking technique and recent emerging precision spectroscopic detection methods.

5.1. Wavelength extension into mid-IR region

Promoted by the demand for spectroscopic applications in mid-IR, dual-comb spectroscopy with a single dual-comb laser with extended wavelength coverage into mid-IR exhibits great potential for the detection of molecular finger-print spectroscopic feature.

Spectroscopic applications of dual-comb lasers toward mid-IR are still rare, as only limited dual-comb laser sources have been developed [31, 70, 126, 127]. As referred in reference [31], the proof-of-principle experiment conducted in our group shows the compatibility of dual-comb spectroscopy in 2 \( \mu \text{m} \) region. For those application in mid-IR, single laser source emitting dual-comb in mid-IR should be developed at first. For solid state lasers, OPOs are still competitive candidates for mid-IR dual-comb spectroscopy [128]. Instead of using two independent mode-locked lasers to drive a single synchronously pumped optical OPO.
Figure 7. (a) Schematic illustration of dual-comb spectroscopy using a wavelength-multiplexing dual-comb laser; (b) comb-resolved spectrum (blue) and the corresponding absorption curve from HITRAN (red); (c) one single comb-line resolved with a resolution of 0.25 Hz (expanding the x-axis of (b) by 10 000 times) [31]. AMP: amplifier; HNLF: highly non-linear fiber; BPF: band pass filter.

[45, 47], a near-infrared dual-comb mode-locked solid state or fiber laser may be utilized as the pump laser. Therefore, a pair of mid-IR optical frequency combs with high tunability may be obtained by using a single dual-comb mode-locked laser pumped OPO. For fiber lasers, the commonly-used silica-based fiber is regarded deficient for laser source above 2 \( \mu \)m. However, chalcogenide fiber, germanate fiber, heavy metal fluoride fiber (ZBLAN, chemically ZrF\(_4\)-BaF\(_2\)-LaF\(_3\)-AlF\(_3\)-NaF), et al, are superior for the transmission in mid-IR [34, 129–131]. For example, optical frequency comb sources based on erbium-ZBLAN have been reported around 2.8 \( \mu \)m and 3.5 \( \mu \)m [132, 133]. There’s no obstacle to design novel mid-IR dual-comb fiber lasers based on the above active fibers. For those on-chip nanophotonics, the realization of dual-comb laser greatly relies on mid-IR pump laser [22, 134].

5.2. Novel precision spectroscopic detection methods

Several competitive high precision spectroscopic approaches are envisioned as follows. Combined with the advances in mid-IR dual-comb lasers, practical and powerful dual-comb mid-IR molecular spectroscopy are expected in the near future.

5.2.1. Intracavity absorption spectroscopy

Intracavity absorption spectroscopy utilizes cavity-enhanced ultra-long interaction length (typically several kilo–meters) to enhance the detection sensitivity, enabling absorptive spectrum measurement of trace-gases with a concentration down to several part-per-million (ppm) or even sub-part-per-billion (sub–ppb) [135, 136]. The combination of intracavity absorption spectroscopy with dual-comb lasers is expected to bring in the practical implementation as well as the ultra-high sensitivity together. However, intracavity
absorption spectroscopy usually suffers from limited detectable spectral range which is determined by gain spectrum of specific laser. In order to overcome this shortage, dual-comb OPOs could be utilized instead.

5.2.2. Multi-dimensional spectroscopy
Apart from the conventional linear spectroscopy, two and more laser beams can interact in the regime of non-linear spectroscopy. For example, an absorption in a sample can be saturated by one strong ‘pump’ beam, thus providing an increased transmission for a weaker ‘probe’ beam. Or, a ‘signal’ beam having a new direction and frequency can be generated in the sample via wave-mixing. More interestingly, when the measurement is made as a function of time instead of frequency, the Fourier-transformed spectra could be functions of multiple time delays and thus multidimensional. With a development over two decades, multidimensional coherent spectroscopy (MDCS) has become an extremely power yet unpractical method. On one hand, a wealth of information hidden from conventional spectroscopic techniques has been successfully unearthed using MDCS method [137–139]. On the other hand, the relative long acquisition time, coarse resolution, and bulky apparatus also set an obstacle for the wide-spread application [140]. To overcome these weaknesses, B Lomsadze et al [141] demonstrated a tri-comb spectroscopic approach that provides a comb-line resolved multidimensional coherent spectrum in 1 s, which is not achievable by any other currently available methods, making comb-based MDCS competitive for atomic spectroscopy (both cold and Doppler broadened), as well as molecular ro-vibrational spectroscopy. The above MDCS experiment still requires tightly phase-locked triple-combs. Recently, several practical multiple-comb generation schemes based on single laser cavity has already been demonstrated [33, 49]. Triple electro-optic-combs have also been utilized for absolute ranging applications [142]. A rapid increase of the appealing research direction on practical multiple-comb generation as well as the related comb-based MDCS and other high precision applications is foreseeable in the near future.

6. Conclusion
To conclude, we have summarized the recent progresses on a distinct dual-soliton generation technique from mode-locked lasers. This unique mode-locking regime draws special attentions in recent years due to the great advantages as a comb source for the powerful dual-comb molecular spectroscopic applications. We present that multiple multiplexing approaches, such as wavelength-multiplexing, polarization-multiplexing, circulation-direction-multiplexing and cavity-space-multiplexing, have been explored for dual-comb generation in various laser sources, including solid state, all-fiber as well as on-chip waveguide laser oscillators. The major advantage of dual-comb operation in a single laser cavity is effective common mode noise cancellation, thus splendid stability between the two combs is feasible. A number of these laser sources have already been successfully applied for dual-comb spectroscopy, resulting in comb-line resolved dual-comb spectroscopy with an extremely simple setup. Their performance is fundamentally limited by ASE noise that is uncorrelated and cannot be removed by sharing one laser cavity. Till now, dual-comb spectroscopy has already been demonstrated close to 2 µm wavelength based on a single free running passively mode-locked thulium-doped fiber lasers. There is no obstacle to extend the wavelength towards the more attractive molecular fingerprint range given that special mid-IR gain medium are available. Driven by the emerging MDCS applications, single laser based multiple comb generation techniques, particularly, in mid-IR, foresees a rapid progress in the near future.

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