

Dual-comb spectroscopy

Nathalie Picqué ^{1,2,3}✉ & Theodor W. Hänsch ^{2,4}

Abstract

Broadband spectroscopy combines wide spectral coverage with high resolution and accuracy, requirements that are often difficult to satisfy simultaneously using conventional approaches. Dual-comb spectroscopy addresses this challenge by using two optical frequency combs with slightly different repetition frequencies to map optical spectra directly into the radio-frequency domain. The method relies on time-domain interferometry and avoids mechanical scanning, enabling precise, rapid and broadband measurements. In principle, the achievable spectral resolution is set by the comb line spacing, defined by the laser repetition frequency. Because the measurement does not rely on geometrical constraints, the dual-comb interferometer offers a conceptual route towards broadband spectroscopy with resolution determined purely by temporal coherence. Over the past two decades, dual-comb spectroscopy has been implemented across the electromagnetic spectrum, from the terahertz to the visible, with ongoing efforts towards the ultraviolet. In this Primer, we present the fundamental principles of the technique, analyse the parameters governing its performance – including resolution, accuracy, signal-to-noise ratio and acquisition speed – and describe practical approaches to data acquisition and processing. We conclude by examining representative applications, current limitations and emerging directions for further development.

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¹Max-Born-Institute for Nonlinear Optics and Short-Pulse Spectroscopy, Berlin, Germany. ²Max-Planck Institute of Quantum Optics, Garching, Germany. ³Department of Physics, Humboldt University of Berlin, Berlin, Germany.

⁴Faculty of Physics, Ludwig-Maximilian University of Munich, München, Germany. ✉e-mail: nathalie.picque@mbi-berlin.de

Introduction

A frequency comb is a spectrum of phase-coherent, evenly spaced narrow laser lines¹. The advent of frequency combs in the late 1990s has profoundly transformed the domain of time and frequency metrology. Frequency combs have provided rulers in frequency space able to measure substantial optical frequency differences and that seamlessly connect microwave and optical frequencies². These developments have been driven, in their initial stages, by the challenges associated with the precise laser spectroscopy of atomic hydrogen and related tests of fundamental physics laws. Another motivating goal has been the development of a clockwork mechanism for optical atomic clocks. In recent years, advances in frequency-comb technology have given rise to new applications and discoveries^{3–8}.

Frequency-comb spectroscopy involves the use of a frequency comb to directly excite or interrogate the sample. The field has seen a surge in interest over the past decade. In this rapidly advancing area and in its emerging applications, frequency combs offer a set of new tools for spectroscopy over broad spectral bandwidths in all phases of matter, a subject of particular interest to molecular spectroscopy. The use of frequency combs has resulted in a substantial enhancement to the performance of state-of-the-art spectrometers, including scanning Michelson Fourier-transform interferometers^{9,10} and dispersers¹¹. For readers interested in these alternative methods, we provide references for further exploration^{12–15}. Importantly, frequency combs have enabled the advent of a new generation of instruments, namely dual-comb spectrometers^{16–21}.

Dual-comb spectroscopy has rapidly matured over the past decade, offering a versatile approach to high-resolution spectroscopy across broad spectral ranges. By using two mutually coherent frequency combs, it can capture entire spectra simultaneously, making it suitable for probing complex molecular structures, detecting weak signals and monitoring fast or transient phenomena. Beyond the laboratory, dual-comb instruments are being developed for environmental sensing, chemical analysis and biomedical diagnostics, including compact, portable systems capable of real-time measurements. Although these capabilities have already transformed spectroscopy, the full potential of dual-comb techniques – such as the possibility of achieving unlimited spectral spans and resolutions – has yet to be realized, offering exciting opportunities for those entering the field.

This Primer provides a tutorial to dual-comb spectroscopy, focusing on the concepts and the physical insights underlying the technique. We review the main ways of implementing a dual-comb spectrometer mostly in linear spectroscopy and of calculating the spectra. We highlight the different comb generators that have been harnessed and their specific features for dual-comb spectroscopy. We summarize the applications that have been explored so far. We conclude by presenting our vision of the prospects, opportunities and challenges. Dual-comb spectroscopy requires that an interferometer is formed with the two frequency combs, and although dual-comb interferometry has also found applications in areas such as distance measurements^{22,23} and holography²⁴, these topics are outside the scope of this Primer. Instead, we concentrate on the most widespread application of dual-comb interferometry: dual-comb spectroscopy.

Experimentation

Principles of dual-comb spectroscopy

In dual-comb spectroscopy, two frequency-comb generators (Fig. 1a) emit trains of pulses at slightly different repetition frequencies, f_{rep} and $f_{\text{rep}} + \Delta f_{\text{rep}}$. We first consider the simplest experiments of linear

absorption spectroscopy, where the beam of one comb interrogates a sample (Fig. 1b). The comb beam that is transmitted through the sample carries an imprint of the sample's transitions. The beams of the two combs are combined on a beam splitter, and their interference is measured on a fast photodetector as a function of time. In the time domain (Fig. 1c), the periodic waveforms of the emission of the interrogating comb followed by the free induction decay of the absorbing sample are oscillating too fast to be directly monitored. Owing to the asynchronism of the two comb lasers, the pulses from the second laser walk through the pulses from the interrogating laser, with a time separation that automatically increments from pulse pair to pulse pair by an amount equal to $\Delta f_{\text{rep}}/(f_{\text{rep}}(f_{\text{rep}} + \Delta f_{\text{rep}}))$ approximately equal to $\Delta f_{\text{rep}}/f_{\text{rep}}^2$. This way, optical delays from $-1/(2f_{\text{rep}})$ to $1/(2f_{\text{rep}})$ are repetitively scanned without moving parts, at a period of $1/\Delta f_{\text{rep}}$. Akin to a sampling oscilloscope, the periodic optical waveforms are stretched in time by a factor $f_{\text{rep}}/\Delta f_{\text{rep}}$. The time-domain interference signal is called an interferogram, and the fraction of an interferogram scanning delays from $-1/(2f_{\text{rep}})$ to $1/(2f_{\text{rep}})$ is known as an individual interferogram.

Experimentally, the signal of the photodetector is often electronically low-pass filtered to suppress the pulse repetition frequency, f_{rep} . The time-stretched waveform therefore appears as a continuous electronic signal. This signal is digitized at a constant clock rate determined by the data acquisition board of the computer. For ideally stable frequency combs, the interferogram thus sampled can be Fourier-transformed to reveal the signal spectrum. In the frequency domain (Fig. 1d), the frequencies of the optical lines of the two combs may be expressed as $f_n = n f_{\text{rep}} + f_0$ and $f'_n = n(f_{\text{rep}} + \Delta f_{\text{rep}}) + f_0 + \Delta f_0$, where n is the integer number of the optical line. The lines of the interrogating comb are attenuated and phase-shifted when in resonance with the transitions of the atoms or molecules. Pairs of optical comb lines, one from each comb, produce beat notes on the detector, forming a frequency comb of line spacing Δf_{rep} . Optical frequencies $n f_{\text{rep}} + f_0$ are thus down-converted into frequencies $n \Delta f_{\text{rep}} + \Delta f_0$. In general, the down-conversion, with a factor $\Delta f_{\text{rep}}/f_{\text{rep}}$, maps the down-converted comb in the radio-frequency domain where signals can be directly digitized and processed with electronics. For a one-to-one correspondence between lines in the optical and radio-frequency domains (Fig. 2a), the sampling conditions imply that the free spectral range in dual-comb spectroscopy is $f_{\text{rep}}/2$. Therefore, Δf_{rep} should be chosen such that

$$\Delta f_{\text{rep}} \leq \frac{f_{\text{rep}}}{2N}, \quad (1)$$

where N is the number of comb lines in the optical spectrum. The optical span of the combs, ΔF , is $\Delta F = N f_{\text{rep}}$. Equation (1) can be similarly expressed as $\Delta F \leq f_{\text{rep}}^2/(2\Delta f_{\text{rep}})$. Beat notes between neighbouring pairs of optical comb lines are imaged in the range $[0, f_{\text{rep}}/2]$, whereas beat notes between non-adjacent pairs are mapped into higher order aliases of the free spectral range $[M f_{\text{rep}}/2, (M+1) f_{\text{rep}}/2]$, where M is an integer index that indicates the aliasing order. If the frequency bandwidth of the photodetector is large enough, a replica of the spectrum, arising from beat notes between increasingly non-adjacent comb lines, occur in the aliases of the free spectral range (Fig. 2b). Strategies for using lasers of incommensurate repetition frequencies or working in aliasing conditions have been discussed but are not widespread. Upon digitalization, the time-domain interference signal is sampled. The Nyquist condition would impose that the frequency bandwidth and sampling rate of the digitizer is higher than the free spectral range $f_{\text{rep}}/2$. However, if the actual spectral span does not cover the entire free spectral range

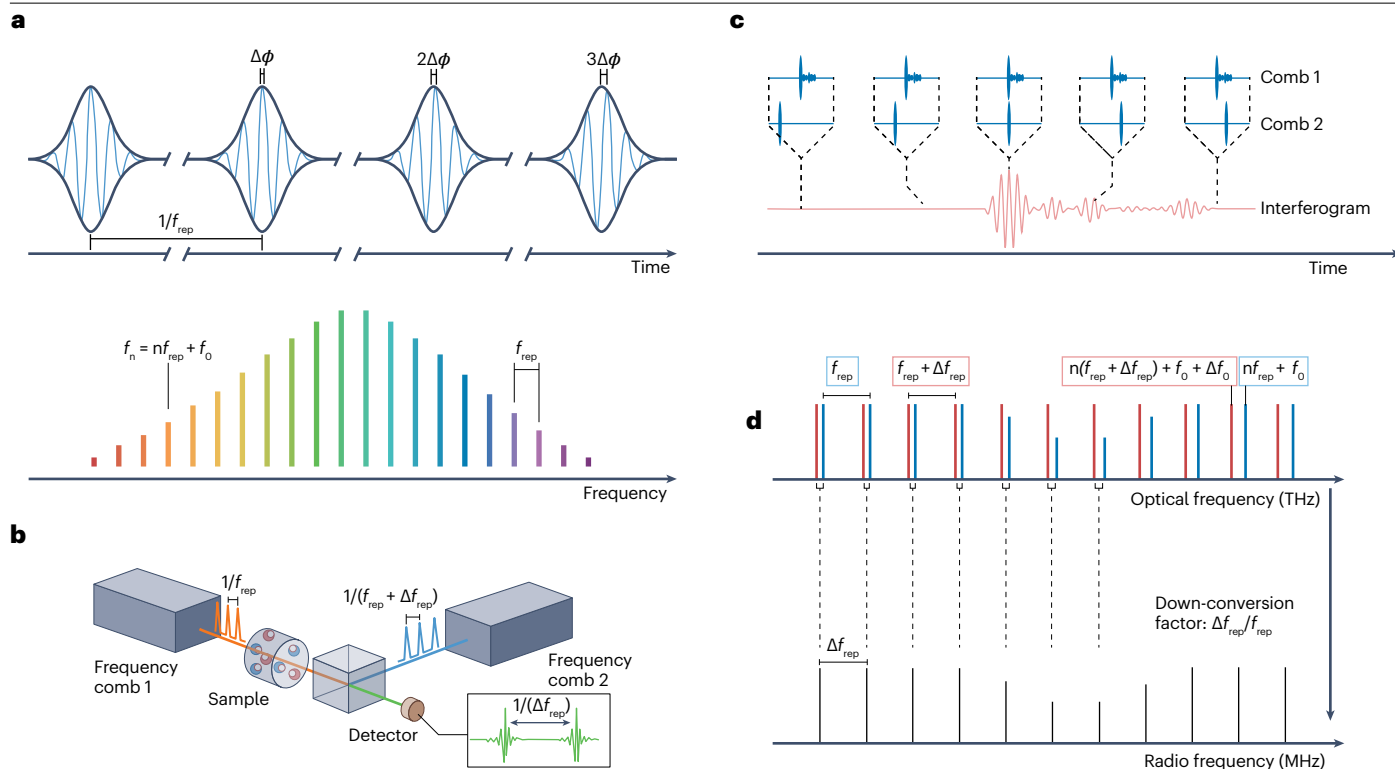


Fig. 1 | Optical frequency-comb and dual-comb spectroscopy. **a**, Time-domain and frequency-domain representations of a frequency comb. A time-domain representation of a train of ultrashort pulses with a period of $1/f_{\text{rep}}$ at the output of a mode-locked laser, alongside the corresponding spectrum of narrow lines of a frequency comb, where f_{rep} is the repetition frequency. The phase shift ($\Delta\phi$) of the carrier wave relative to the pulse envelope induces a translation ($f_0 = f_{\text{rep}} \Delta\phi/2\pi$) of all comb lines from harmonic frequencies (nf_{rep}), where n is the integer number of the optical line. **b**, In a dual-comb spectrometer, two frequency-comb generators of slightly different repetition frequencies are used. In the simplest

configuration, the beam of one comb is transmitted through an absorbing sample contained in a cell and is combined with the beam of the second comb on a beam splitter. The two beams interfere at a fast photodetector and the time-domain interference is measured as a function of time. **c**, In the time domain, the pulses from the second comb walk through the waveforms of the first comb and optically sample the waveform with a time delay that automatically increases from pulse pair to pulse pair. **d**, In the frequency domain, the beat notes between pairs of optical comb lines, one from each comb, generate a down-converted radio-frequency comb, which can be measured using digital electronics.

$f_{\text{rep}}/2$, under sampling may be performed by carefully choosing a faster sampling rate (Fig. 2c).

In the dispersive configuration (Fig. 1b), one comb interrogates the sample and the other acts as a local oscillator, with the complex spectrum revealing the spectral response of the narrow laser lines of the interrogating comb alone. The amplitude spectrum shows the absorption experienced by the sample and the phase spectrum shows the dispersion. This configuration is ideal for precise investigations of spectral line shapes. Alternatively, it is also possible to position the absorbing sample after the beam combiner. This enhances the sensitivity to absorption because both comb beams experience attenuation by the absorber. However, this arrangement does not provide any dispersion information, and each spectral element in the dual-comb spectrum reports on the absorption experienced by two comb lines at two different frequencies.

As a technique of Fourier-transform spectroscopy, dual-comb spectroscopy is a multiplex technique as all the spectral elements are simultaneously recorded on a single photodetector. This is different for dispersive spectrometers, such as grating spectrographs, which rely on sequential or on multichannel parallel recording. The simultaneous recording allows exceptional consistency of the spectral data.

Systematic effects such as changes in the sample affect the spectrum in the same way across the entire spectral bandwidth. Even where there is no gain in recording time, the experimental time may be shorter since there is not any need for tuning and calibrating the frequency of the light sources, especially for large span. The use of a single photodetector enables operation in any region where frequency combs are available. Replacing the single photodetector by a detector array or matrix enables additional features such as spatial resolution. Initially, the multiplex advantage was expressed as an advantage in signal-to-noise ratio in situations where detector noise dominates. Detector noise is generally not so important with the intense coherent light used dual-comb spectroscopy.

Resolution in dual-comb spectroscopy

The dual-comb interferometer is fundamentally different from any other spectrometer over broad spectral bandwidth, including other instruments of frequency-comb spectroscopy. Indeed, the dual-comb interferometer performs direct time or frequency measurements. Dispersive or interferential instruments such as a grating spectrometer or a Michelson interferometer, rely on wavelength measurements. The resolving power, R , is the ratio of the maximum path difference,

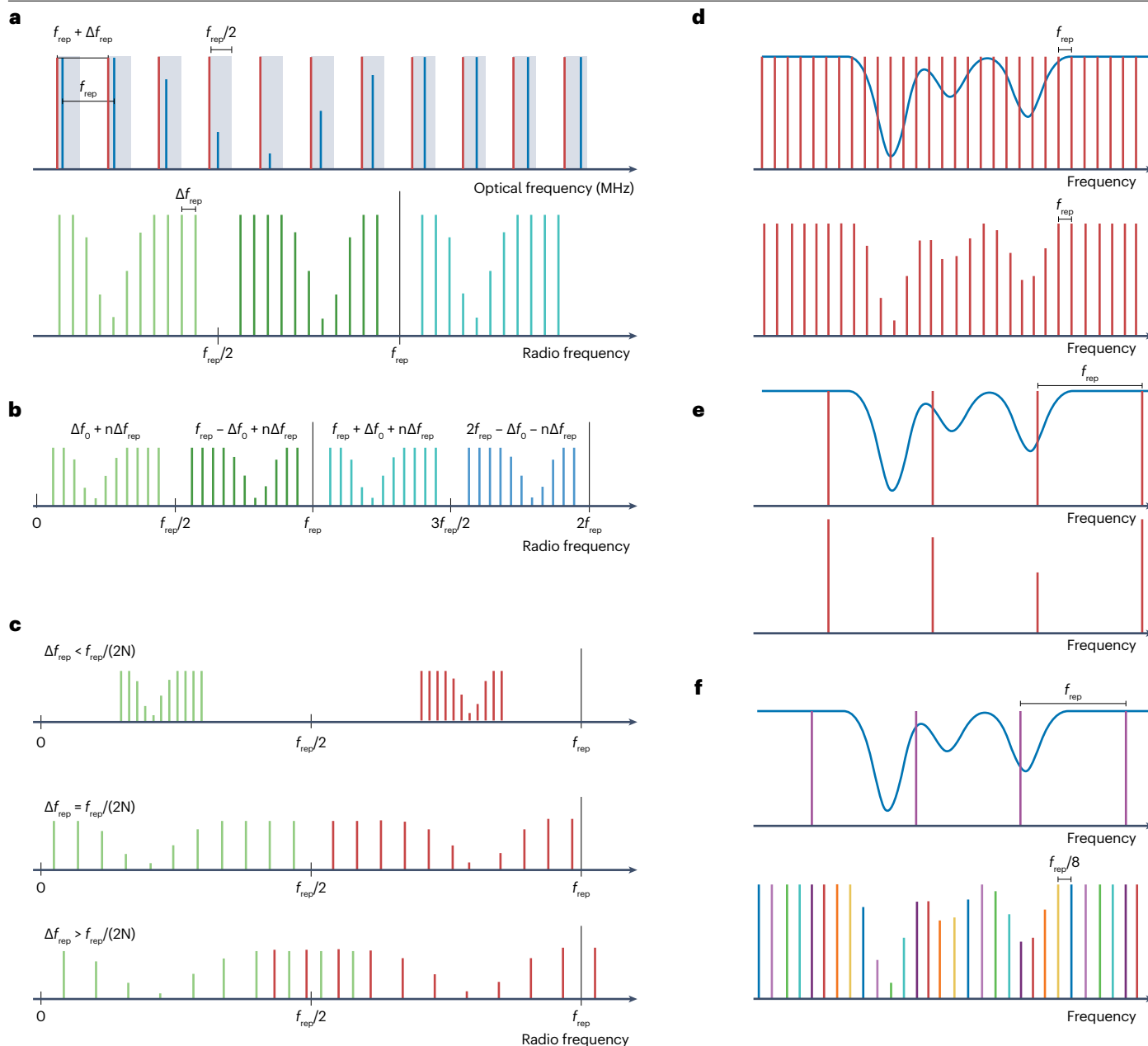


Fig. 2 | Aliasing, sampling and interleaving in dual-comb spectroscopy. **a**, To avoid aliasing, the beat notes between pairs of adjacent comb lines need to be kept below the repetition frequency, $f_{\text{rep}}/2$. **b**, The beat notes between pairs of non-adjacent comb lines create aliases over replica of the free spectral range, $[nf_{\text{rep}}/2, (n+1)f_{\text{rep}}/2]$, where n is the integer number of the optical line. **c**, Choosing appropriately Δf_{rep} avoids aliasing. N is the number of comb lines in the optical spectrum **d**, Dual-comb spectroscopy is the only technique able to

measure at any resolution beyond the limits owing to geometry. Adapting the comb repetition frequency, for instance by pulse picking, enables measurement of the entire spectrum in a single recording. **e**, The spectrum is sampled by the discrete comb lines; thus, their line spacing, f_{rep} , defines the resolution in a single spectrum. **f**, By tuning the frequency of the lines of the interrogating comb and recording as many spectra as there are tuning steps, the resolution can be improved by interleaving.

Δ , to the wavelength, λ , hence high resolution requires bulky instruments, and resolution is fundamentally limited by wavefront distortions. With a dual-comb interferometer – which can be viewed as a multi-heterodyne device – the resolving power becomes the ratio of the maximum optical retardation to the period of the optical wave. Dual-comb spectroscopy is therefore the only technique that can, for

any spans and any spacing, potentially reach a resolution equal to the comb line spacing, freed from geometric limitations and aberrations. In theory, the resolution is only limited by the uncertainty principle, which is in practice quite generous. This apparently simple yet fundamental distinction, first highlighted in refs. 12,25, has not yet been widely exploited in experiments that would advance the state of the art²⁶. Using

at least one comb with a low-repetition frequency that matches the desired resolution enables spectrum acquisition in a single recording²⁶ (Fig. 2d). This remarkable feature of dual-comb spectroscopy allows to work beyond the limitations imposed by wavefront distortions in traditional spectrometers and achieve much higher resolutions than existing spectrometers for broadband light sources²⁶.

The resolution depends on the excursion of optical delays in the interferogram. If the interferogram is sampled or if a portion of interferogram is Fourier transformed, over a range of delays shorter than $[-1/(2f_{\text{rep}}), 1/(2f_{\text{rep}})]$, the spectra are convolved by an instrumental line shape primarily defined by the measurement time window, called the apodization window. The resulting instrumental line shape has a width of similar magnitude to that of the atomic or molecular resonances being investigated. Therefore, retrieving line parameters requires nonlinear least-squares fitting to account for convolution by the instrumental line shape, reducing precision and accuracy. So far, a large proportion of experiments of dual-comb spectroscopy have been performed in this apodization regime and do not reach a resolution able to resolve individual comb lines. However, even in ordinary Fourier-transform spectroscopy, the use of laser-like coherent radiation offers some important advantages over the commonly used incoherent light sources. The measurement speed may be improved by several orders of magnitude for the same signal-to-noise ratio and resolution⁹. Laser beams allow multi-pass cells or high-finesse resonators to be used to enhance sensitivity to weak absorptions of species such as trace gases, as well as the use of detectors of low sensitivity. Laser beams are also advantageous for a wide range of sample configurations; for instance, they can interrogate outdoor open-air paths of a few kilometres in length or eliminate background noise from the focal plane in microscopy.

The dual-comb interferometer is far more than a Fourier-transform spectrometer without moving parts. In the dispersive configuration, when only one comb interacts with an absorbing sample before interfering with a local oscillator, the spectrum is sampled by the lines evenly spaced by f_{rep} of the probing comb. With a dual-comb interferometer, by construction, the optical delay is at maximum $1/(2f_{\text{rep}})$, leading to a nominal resolution of f_{rep} , which is the comb line spacing. A maximum optical delay of $1/(2f_{\text{rep}})$ is sufficient to resolve the spectral response of each comb line. In a single interferometric measurement, no spectral features sharper than the comb line spacing can be observed in the dual-comb spectrum (Fig. 2e). Following Rayleigh criterion, no spectral features narrower than $2.4f_{\text{rep}}$ can be separated. In other words, the nominal resolution in a dual-comb spectrum resulting from a single acquisition sequence is at best equal to the comb line spacing. Akin to tunable laser spectroscopy, the width of an optical comb line, which can be very narrow, determines how precisely the spectral sample is defined. The precision with which the spectral samples are defined may thus be much better than the resolution and, if the width of the comb lines is much narrower than the spectral transitions, the instrumental line shape of the spectrometer may be negligible relative to the spectral profiles, such as the Doppler-broadened lines of small molecules at room temperature. We note that the resolution in a spectrometer is its ability to distinguish spectral features such as molecular or atomic lines, thus the width of the optical comb lines, although an interesting parameter to quote, does not define the spectral resolution in a single acquisition sequence.

Increasing the resolution requires the use of a comb with a smaller line spacing²⁶ (Fig. 2d). However, this is not what is currently used as resolutions better than the comb line spacing have

been obtained by interleaving sets of independent spectra¹⁸ (Fig. 2f). Most frequency-comb generators can be precisely controlled, making it possible to measure a sequence of independent spectra where the position of the optical lines of the interrogating comb $f_n = nf_{\text{rep}} + f_0$ are changed stepwise in a predefined manner from one recording to the next¹⁸. For example, the carrier-envelope offset frequency can be tuned. At each step of the N steps, the time-domain interference is recorded so that as many spectra as there are steps are recorded. A posteriori stitching together the independent spectra improves the resolution to f_{rep}/N , assuming that the steps are evenly spaced in the frequency range (Fig. 2f). Ultimately, the resolution using interleaving is limited by the width of the optical lines of the probing comb.

Measuring interference

Here, we consider the stability requirements that need to be achieved to measure dual-comb spectra. A dual-comb spectrometer is an interferometer, therefore conditions where time-domain interference between the two combs is observed need to be met. In the simplest configuration of linear absorption spectroscopy, the two combs should be very similar except for their slightly different line spacing. This requirement applies to spectral span, power spectral density and polarization. In any experimental setting, the pulse train of a comb generator experiences timing and phase jitters that deviate the experimental waveforms from the theoretically periodic ones. Relative fluctuations between the two combs can cause artefacts and degrade the resolution and precision. The absolute stability of the interrogating comb and the mutual coherence of the two combs are two crucial, independent requirements that can greatly affect the quality of the spectra.

The absolute frequency stability of the interrogating comb is crucial for realizing the benefits of dual-comb spectroscopy in practice. Drift in the comb lines affects the spectral profiles, resolution and accuracy. The sample is periodically excited by periodic coherent waveforms and thus interrogated by narrow, equidistant laser lines. The sharpness and stability of these comb lines determine how precisely the spectral samples are defined. If the lines of the interrogating comb are narrow, the instrumental line shape may be negligible compared with the width of atomic and molecular transitions. Furthermore, if the interrogating comb is referenced to a radio-frequency clock, the frequency scale of the dual-comb spectra can be calibrated directly against it. The choice of stabilization and referencing scheme depends on the scientific objectives. For instance, a comb referenced to a radio-frequency clock using the self-referencing scheme may exhibit lines with a width of a few hundred kilohertz. This is narrow compared with the Doppler width of most small, light molecules in the infrared, visible or ultraviolet (UV) spectrum, which is typically hundreds of megahertz or more. Therefore, stabilizing the interrogating comb to a radio-frequency clock is sufficient for most spectroscopic endeavours explored with dual-comb spectroscopy thus far. Narrower comb lines can be obtained by stabilizing a comb against an optical narrow-linewidth continuous-wave laser². With some additional steps, absolute calibration of the frequency scale can also be achieved. An alternative to stabilization is to track the drifts of unstabilized, free-running combs. However, if the instabilities are commensurate with the width of the transitions of atomic or molecular samples, distortions may be present in the resolution and line shapes. Monitoring these instabilities may also result in a more cumbersome set-up than directly stabilizing the comb, a process that now follows standard techniques and is commercially available as a whole or in parts.

An independent requirement for artefact-free spectra is that the two frequency combs must be mutually coherent over the measurement time. Spectral line shapes, resolution and increase of the signal-to-noise ratio with measurement time are compromised when mutual coherence is not enforced. Long measurement times are the key to achieving a resolution equal to the comb line spacing and the highest signal-to-noise ratio. This means that the relative fluctuations of the two combs must be kept within a fraction of the wavelength for each interferometric sample. In the time domain, residual instabilities of the frequency combs, even if they benefit from state-of-the-art stabilization, induce variations in the time intervals between the excitation and sampling pulses, which are indeed subject to some variations δt , which appear in the detector signal stretched in time by the factor $f_{\text{rep}}/\Delta f_{\text{rep}}$. The slippage of the carrier phase relative to the pulse envelope arises from dispersion in the laser cavity. If the carrier-envelope phase-shift frequencies of the two combs differ by Δf_0 , the relative phases of the pump and probe pulses will change by an additional $2\pi\Delta f_0/f_{\text{rep}}$ between two interferometric samples. All frequencies in the Fourier spectrum of the detector signal will also shift by Δf_0 . Provided that Δf_0 remains constant, the spectral translation can be accounted for when Δf_0 is measured²⁷. In the frequency domain, heterodyne beat notes produced by pairs of comb lines, one from each comb, create a radio-frequency comb. Assuming that the residual relative frequency fluctuations between the two optical combs are uncorrelated, the lines of the radio-frequency comb generally have a width equal to the square root of the sum of the squared widths of the optical comb lines²⁸. Meanwhile, their line spacing is reduced by several orders of magnitude to Δf_{rep} . Over the measurement time, the width of the radio-frequency comb lines needs to be narrower than Δf_{rep} for benefiting from the full resolution capabilities. Otherwise, the resolution is compromised, which may induce artefacts and misestimations of any attempt for quantitative spectroscopy. In addition, for the signal-to-noise ratio to increase as the square root of the measurement time, the intrinsic width of the beat note should be narrower than that induced by the finite measurement time, which is inversely proportional to the measurement time. Importantly, the observed beat note width characterizes the relative coherence of the two combs, but it is insufficient to quantify either the precision of the spectral samples or the spectral resolution. For example, even with two unstabilized combs whose relative fluctuations are fully correlated, narrow radio-frequency beat notes may still be observed, whereas the spectral resolution is degraded by absolute drifts of the comb lines. For broad-span combs, Δf_{rep} can be less than a few hundred hertz, and the measurement time required to reveal a broad spectrum with a signal-to-noise ratio of several hundred over the span can be longer than several minutes under conditions where systematic effects are well controlled and minimized. The requirement for mutual coherence between the combs has been therefore challenging to tackle, as it involves achieving beat notes between pairs of comb lines of a sub-hertz linewidth or even less.

The frequency comb used as a frequency ruler in metrology and comb-assisted spectroscopy is based on stabilizing the pulse repetition frequency and the carrier-envelope offset frequency against a radio-frequency clock using the self-referencing $f-2f$ scheme. In this case, the repetition frequency and the carrier-envelope offset frequency are controlled by slow feedback loops with a bandwidth of a few kilohertz, operating over short timescales and utilizing the flywheel stability of the laser oscillator. Such combs are accurate for frequency metrology and dual-comb spectroscopy. However, as the width of their lines is on the order of several hundred kilohertz, two such combs are

not sufficiently stable to enable dual-comb spectroscopy²⁷. Table 1 provides a simplified summary of experimental configurations and typical performance.

Mutual coherence, detection and averaging

Several powerful solutions are now available to achieve mutual coherence, making it effectively enforced during experiments. For instance, locking two lines of each comb to a pair of narrow-linewidth continuous-wave lasers has produced a coherence time that is inversely proportional to the linewidth of the continuous-wave lasers^{19,29}. This scheme enables continuous time-domain measurements to be taken over a period of approximately 1 s. Phase-correction techniques derived from Fourier-transform spectroscopy can then be used to efficiently average many 1-s recordings and improve signal-to-noise ratio. Through the use of feed-forward relative stabilization of the difference in offset frequencies^{30,31}, it has been possible to directly average interferograms over a period of 1 hour without any corrections. This suggests that the experimental phase control of the dual-comb interferometer can be arbitrarily long, facilitating broadband frequency metrology and detection in light-starved conditions. Another powerful approach uses analogue^{27,32} or digital processing³³ to correct the detected interferometric signal for relative fluctuations between the two combs in real time, or using a posteriori corrections^{34,35}. Such schemes have even been implemented with free-running or loosely locked lasers. These schemes also correct for residual fluctuations in stabilized systems and are most effective when implemented alongside stabilized systems, as the bandwidth of stabilization electronics is generally not as broad as the frequency bandwidth of interferometric signals. A third, increasingly common approach involves improving the mutual coherence of the dual-comb sources by design^{36,37}. If the two laser sources share optical or optoelectronic components, the relative fluctuations in dual-comb systems can be much lower than twice those of a comb source in an uncorrelated system. In some configurations involving electro-optic modulators, the mutual coherence time exceeds 1 s (refs. 38,39). Nevertheless, such designs are generally not sufficient to avoid active stabilization or digital corrections. Although common noise rejection has been demonstrated^{36–39}, these set-ups in general do not suppress sufficiently all instabilities to avoid some type of stabilization or a posteriori correction. Many of these solutions have not yet been thoroughly benchmarked, for example, by comparing the retrieved line parameters with those measured by state-of-the-art spectrometric techniques, or by carefully analysing the systematic effects.

Dual-comb spectroscopy has so far mostly been used for linear absorption spectroscopy. In such a case, the dual-comb time-domain interferogram is detected on a fast photodetector, which detects radiation at the same frequencies as the transitions of the sample, although some demonstrations have investigated schemes of up-conversion^{40–43}. The photodetector should have a bandwidth able to sample the waveforms in the interferogram, whose maximum down-converted frequency is $N\Delta f_{\text{rep}}$. For the fastest measurements, Δf_{rep} is set at the Nyquist limit and the photodetector has an electronic bandwidth of $f_{\text{rep}}/2$. In the dispersive configuration, the two outputs of the interferometer present similar unmodulated backgrounds but opposite phases for the interferometric signal. They may be detected with similar photodetectors and their electric response may be subtracted for higher signal-to-noise ratio and nonlinearities minimization⁴⁴. The electric signal, possibly filtered and amplified, is subsequently digitized by a fast digitizer. Nonlinearities in the detection chain generate systematic effects, which can be very subtle and affect the precise determination of spectral line

Table 1 | Possible stabilization configurations for dual-comb spectrometers and associated performance

Attribute	Optical lock ¹³³	Radio-frequency lock and relative lock for mutual coherence ³⁰	Radio-frequency lock	Free-running with high mutual coherence ²⁷	Free-running ²⁰
Stabilization technique	Stabilization of each comb to two ultra stable continuous-wave lasers	Full stabilization of at least one comb to a radio-frequency clock (radio-frequency lock)	Radio-frequency lock	None	None
Mutual coherence scheme	Directly derives from the stabilization	Relative stabilization, for example through feed-forward scheme	None	Relative stabilization or adaptive sampling	None
Width of optical comb lines	<1 Hz	<100 kHz	<100 kHz	>10 MHz	>10 MHz
Width of heterodyne beat note between comb lines	<1 Hz	<1 Hz	<100 kHz	<10 Hz	>100 s MHz
Typical characteristics of dual-comb spectra					
Spectral resolution without interleaving	Comb line spacing f_{rep}	Comb line spacing f_{rep}	>1 GHz	Comb line spacing f_{rep}	>1 GHz
Precision of spectral sampling	<100 Hz	<1 MHz	>GHz	>10 MHz	>GHz
Averaging times	>1 s	>100 s	<100 μ s	<10 s	<100 μ s
Potential quality of spectroscopic data	Excellent	Excellent	Poor	Fair	Poor

The numbers represent simplified examples and performance reported in dual-comb spectroscopy experiments but do not cover all specific implementations. f_{rep} is the repetition frequency. NA, not applicable.

parameters. The fast sampling rate of the digitizer, often higher than several tens of millions of samples per second, generates challenges for reaching a high dynamic range and produces considerable volumes of data. Real-time digital signal processing facilitates the handling of this data output.

The interferogram recurs at a period of $1/\Delta f_{\text{rep}}$, which provides the shortest acquisition time for comb line resolution. The purpose of acquiring interferometric time sequences longer than $1/\Delta f_{\text{rep}}$ is to improve the signal-to-noise ratio, which generally scales with the inverse of the number of comb lines (Box 1). A single interferogram usually does not lead to a sufficient signal-to-noise ratio, so efficient averaging is crucial. Direct averaging of interferograms in the time domain saves considerable computing time and is possible under specific comb parameter and sampling conditions. In the time-domain, the interferometric optical sampling generates time-stretched waveforms where the waveform envelope has a periodicity of $1/\Delta f_{\text{rep}}$ but a waveform-to-waveform phase shift of $\Delta\phi = 2\pi\Delta f_o/\Delta f_{\text{rep}}$ is experienced. In the frequency domain, the beat notes between pairs of optical comb lines generate a radio-frequency comb with a line spacing of Δf_{rep} and carrier-envelope offset frequency Δf_o . Akin to any frequency comb, a periodicity of $1/\Delta f_{\text{rep}}$ of the time-domain interferometric waveform is only achieved if $\Delta\phi = 0$ (modulo 2π), which corresponds to $\Delta f_o = 0$ (modulo Δf_{rep}). Moreover, the digitization of the interferogram must be performed in reproducible sampling conditions³⁰. For example, all electronics can be referenced to the same radio-frequency clock, and the digitization rate can be an integer multiple of the difference in repetition frequencies Δf_{rep} . An alternative to extensive averaging is to reduce Δf_{rep} and acquire fewer interferograms at a lower sampling rate with an adapted integration time per sample. Depending on the noise figures, this can lead to a higher signal-to-noise ratio in the same measurement time, as slower digitizers with a higher dynamic range can be used. This also results in smaller data files.

Nonlinear spectroscopy

Although dual-comb spectroscopy has primarily been used for linear spectroscopy of small molecules, a frequency comb is often generated

by an ultrashort-pulse mode-locked laser. The high energy of the pulses can be harnessed for creating nonlinear phenomena at the sample, enabling nonlinear spectroscopy over broad spectral bandwidth and for multidimensional spectroscopy without delay lines. Starting with nonlinear Raman^{45–49} and two-photon excitation spectroscopy^{50–52} (Box 2), a variety of schemes have been demonstrated, other variations of pump-probe⁵³, four-wave mixing⁵⁴ and two-dimensional spectroscopy^{55,56}. Applications to transient absorption and various schemes of asynchronous sampling have also been reported^{57,58}.

Unlocking the full potential of nonlinear spectroscopy with two or more frequency combs still requires overcoming technical challenges. Compared with linear dual-comb spectroscopy, schemes of nonlinear dual-comb spectroscopy bring additional complexity. Although frequency combs of a high repetition frequency (on the order of 50–300 MHz) provide the most reliable and developed technology for dual-comb spectroscopy, such laser systems only provide a pulse energy on the order of a few nanojoules at most, even when amplified, making the efficient nonlinear excitation of gas-phase molecules challenging. On the other hand, the high repetition frequency can create local heating and damage in the condensed phase. The excitation probability and the spectral resolution require trade-offs. For coherent excitation, a high repetition frequency compensates for the associated low pulse intensity. Conversely, for short coherence time, the mismatch between the optical-delay scan in the dual-comb system and the coherence time may generate dead times during which no useful signal is measured, meaning a poor measurement duty cycle. For samples in the condensed phase, the duty signal is often lower than 1%⁴⁶. Ultrashort pulses or pulses with a well-defined chirp must often be obtained at the sample, requiring careful dispersion management. By contrast, linear spectroscopy is insensitive to chirps, which can be advantageous for, for example, minimizing detector nonlinearities. The nonlinear response of the sample is usually small and may be buried into stronger signatures, requiring additional strategies to distinguish the signal of interest from the much larger background. For example, the interferometric modulation may be encoded into a background-free signal, such as an intensity modulation in excited fluorescence or in population of ions.

Box 1 | Signal-to-noise ratio and quality factor in dual-comb spectroscopy

The expressions describing the signal-to-noise ratio in dual-comb spectroscopy are similar to those in Fourier transform spectroscopy, for which refs. 125,190 provide a good introduction for linking the signal-to-noise ratio in the time domain to that in the spectrum. The use of coherent light sources made of narrow laser lines are the main features specific to dual-comb spectroscopy.

Qualitatively, essential scaling laws are worth consideration. At constant average power onto the detector, the signal-to-noise ratio scales inversely proportional to the number of comb lines¹⁶⁶. In low power conditions such as in photon counting, it scales inversely proportional to the square root of the number of comb lines¹¹⁹. At short measurement times, increase of the signal-to-noise ratio over time requires that mutual coherence between the two combs is achieved or reconstructed. For extensive averaging times, systematic effects, such as parasitic optical fringes, may prevent improvement of the signal-to-noise ratio. Typically, the signal-to-noise ratio in dual-comb spectroscopy has been dominated by phase noise, intensity noise of the laser sources or noise of the detection system. Recently, shot-noise-limited signal-to-noise ratio has been reached in dual-comb spectroscopy¹¹⁹, as well as subshot-noise level using squeezing¹⁹¹. Although reaching the shot-noise limit means that, within the limits of classical physics, the photons are being optimally used, better signal-to-noise ratio can still be obtained in conditions where other noise sources dominate. More detailed discussions and

quantitative derivations may be found in refs. 119,166. A more subtle effect — which is usually not regarded as noise but which creates systematic effects and distortions in the spectra — is nonlinear in the detection system. This effect is also well known in the context of Fourier-transform spectroscopy and the use of ultrashort-pulse lasers in dual-comb spectroscopy makes it particularly relevant.

The signal-to-noise ratio has been reported inconsistently across various publications. It has on occasion been defined as the height of a comb line divided by the standard deviation of the noise floor, providing an overestimation overlooking multiplicative sources of noise. The signal-to-noise ratio is better described when calculated as the inverse standard deviation of the normalized absorption baseline, made of the comb line maxima in spectra with resolved comb lines.

For comparing spectra measured at different laboratories, spectroscopists have traditionally used a quality factor Q , introduced in ref. 192, as $Q = MB_m/N_{\text{eff}}$ where M is the number of spectral elements (the number of comb lines in dual-comb spectra with resolved comb lines), B_m is the mean spectral energy over the free spectral range, and N_{eff} is the standard deviation of the noise. $B_m/N_{\text{eff}} = \text{SN}$ is the average signal-to-noise ratio. Typically, a figure of merit F has been used, which corresponds to this quality factor normalized to unit of measuring time $F = \text{SN} M/T^{1/2}$, where T is the measurement time. For spectra with many comb lines with well-controlled systematic effects, a figure of merit F on the order of $10^7 \text{ Hz}^{1/2}$ has been a good result.

Frequency-comb technology

A dual-comb spectrometer exhibiting all the advantages of the technique — broad spectral coverage in a region of spectroscopic interest, absolute referencing and narrow instrumental line shape — is not yet commercially available as a turnkey system and must be custom built. Dual-comb spectroscopy has in the past decade been demonstrated with most of the available frequency-comb generators and the body of literature provides convincing evidence that the technique can be successfully implemented with any type of frequency-comb synthesizers. In this section, we summarize some properties of the most common frequency-comb generators in the specific context of their applicability to dual-comb spectroscopy. We do not enter into the specificities of every particular design and only sketch general trends.

Frequency-comb generators used for dual-comb spectroscopy usually consist of a primary generator (Fig. 3) that emits in the near-infrared region — a range often less relevant for spectroscopy — followed by secondary methods, such as nonlinear frequency conversion, to access other spectral regions (Fig. 4). Table 2 provides elements of comparisons of the most common frequency-comb generators, whereas Fig. 5 illustrates the spectral extension of dual-comb spectroscopy over the electromagnetic spectrum using these techniques. Integrated optics is expanding the technological landscape for frequency combs, from comb generation to spectral shifting and broadening^{59–61}.

For each of the technologies presented in Table 2, designs where two asynchronous trains of pulses are generated from a single laser oscillator or a device where many optical elements are shared for common noise suppression have been demonstrated. This includes semiconductor disk lasers⁶², microresonators with counter-propagating pumps⁶³, fibre oscillators^{64–67} and solid-state oscillators^{68–70}, including

high-power thin-disk oscillators⁷¹. Asymmetries in the resonator's nonlinear interactions allow distinct modes or propagation states to oscillate with different round-trip times, thereby generating two frequency combs with slightly different repetition frequencies^{63,65}. Other designs facilitate selecting the difference in repetition frequency^{62,67,71}, for example, by designing a laser cavity where two spatially-separated beams are generated using a biprism⁷² or a birefringent plate⁶². In systems based on electro-optic modulators, a number of components can also be shared^{38,73–75} to increase the mutual coherence time up to more than one second. Most of these schemes essentially shift the challenge of stabilizing the two combs to the challenge of building a special dual-comb laser system. Simplification depends on user knowledge, and so far, no novel scientific capabilities have been demonstrated as enabled by these systems. We refer the reader to other publications for experimental details^{36,37,76}.

Fibre laser systems. An ultrafast fibre laser is an ultrashort-pulse laser where the gain medium is a doped fibre, and a major part of the resonator is made of fibre (Fig. 3a). For frequency-comb synthesizers, the most common fibre gain medium is a fibre doped with rare earth ions, in particular erbium for emission at $1.5 \mu\text{m}$ and ytterbium for emission at $1.0 \mu\text{m}$. Passive mode-locking is achieved efficiently, in particular using nonlinear amplifying loop mirrors⁷⁷ or semiconductor saturable absorbers⁷⁸. The alignment-free fibre cavities can include fast actuators such as electro-optic modulators⁷⁹, which control the repetition frequency and the carrier-envelope offset frequency with a frequency bandwidth up to several megahertz, enabling the implementation of fast feedback loops, which are key to low phase-noise operation⁸⁰. The beam quality can be excellent, even at very high power.

Bulk solid-state laser systems. A bulk solid-state laser is an oscillator where the gain medium is a bulk crystal or a glass. Although the overall concept of a bulk mode-locked laser is very similar to that of a fibre laser, some technical differences may orient the choice towards one or another (Fig. 3a). Bulk oscillators enable direct laser emission in spectral regions where fibre lasers are not available. Because they can feature lower gain and lower resonator losses than fibre lasers, bulk lasers without any servo-control system can exhibit currently lower intensity noise and lower phase noise. However, stabilized fibre lasers with fast actuators outperform the phase noise properties of bulk lasers, where the implementation of intracavity fast actuators is not as developed. Intensity noise is also well suppressed by differential detection of the two outputs of the dual-comb interferometer. Bulk solid-state lasers have not been as widely used as fibre lasers for high-quality dual-comb spectroscopy and they have not demonstrated overall superior performance, although some oscillators do present some distinguishing features such as direct emission at longer wavelengths or higher power^{46,81–83}.

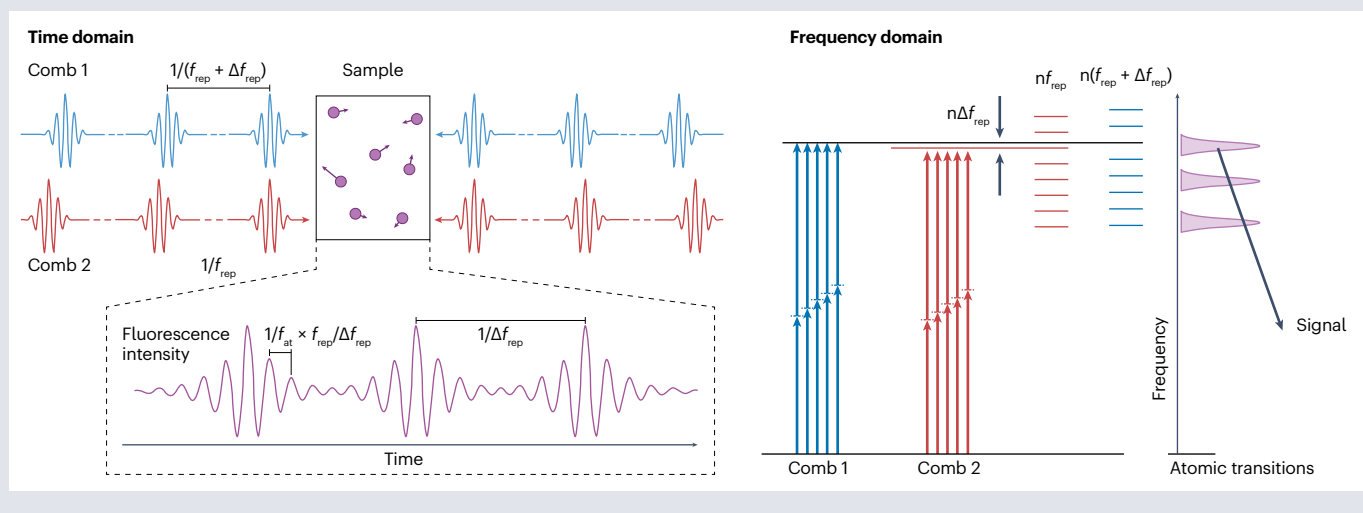
Electro-optic combs. A continuous-wave laser can be modulated in phase and amplitude by a cascade of electro-optic modulators to generate a frequency comb⁸⁴, which can be further broadened a nonlinear fibre³⁸ (Fig. 3b). When no filtering cavity is in line with the set-up, the line spacing and the optical carrier of the frequency comb are freely adjustable over a broad range. However, the phase noise multiplies as one gets away from the continuous-wave carrier quickly creating coherence collapse and therefore the span of such combs is usually limited to less than a thousand lines. Such frequency-agile frequency combs are suited to dual-comb experiments where short measurement times over narrow spans are sought for^{38,73,74,85–87}. Octave-spanning combs using cavity filtering have been demonstrated using complex bulk set-ups⁸⁸. Nanofabricated electro-optic combs using the low-loss thin film lithium niobate platform with or without cavity filtering hold much promise for on-chip dual-comb spectrometers⁸⁹.

Semiconductor lasers. Semiconductor lasers are often electrically pumped and compact. Their development as frequency-comb

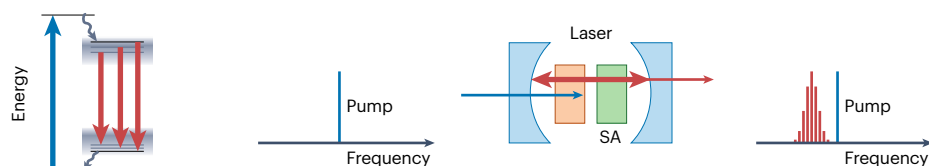
Box 2 | Two-photon excitation dual-comb spectroscopy

We use the example of two-photon excitation spectroscopy, which greatly reduces the Doppler broadening of excited transitions to illustrate the many possibilities enabled by nonlinear dual-comb spectroscopy⁵². Dual-comb two-photon excitation spectroscopy can be understood as an extension of optical Ramsey-interference techniques. In the time domain, two pairs of counter-propagating pulse trains with slightly different repetition frequencies f_{rep} and $f_{\text{rep}} + \Delta f_{\text{rep}}$ form two trains of standing-wave pulses that excite the sample, in this case, an atomic vapour. Initially, atoms are excited into superpositions of coherent states by the first standing-wave pulse, setting the phase origin. An atomic coherence then oscillates at the atomic resonance frequency f_{at} . The second pulse, delayed by a time increment that changes automatically from pulse pair to pulse pair, further excites the atoms, leading to interference between excitations. The upper-state population is modulated as a function of the variable time delay between two pulses from the two asynchronous sources, and this modulation at a frequency of $f_{\text{at}} \times f_{\text{rep}} / \Delta f_{\text{rep}}$ can be observed in the intensity of the atomic fluorescence

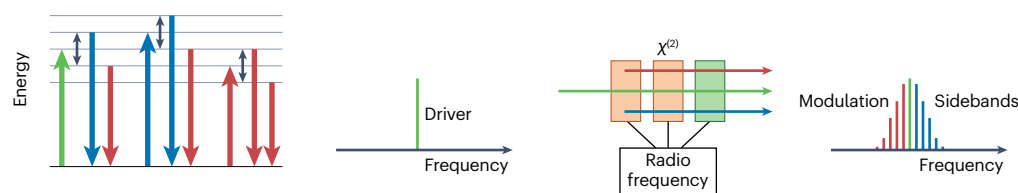
signal, which also decays over time, influenced by, for example, the upper state's lifetime or collision effects. The Fourier transform of this fluorescence signal yields a Doppler-free two-photon excitation spectrum. In the frequency domain, a frequency comb excites two-photon transitions when the sum of comb line frequencies matches atomic line frequencies, which makes the excitation as efficient as with a continuous-wave laser of the same average power^{193–195}. With two frequency combs of slightly different repetition frequencies, the excitation probability includes an interference term oscillating at a distinct frequency. Dual-comb two-photon excitation spectroscopy presents a unique host of features: it is possible to generate combs in regions where continuous-wave lasers are not available as the use of ultrashort pulses enables efficient nonlinear frequency conversion; dual-comb two-photon is background free and Doppler free, and it allows simultaneous observation and unambiguous identification of multiple transitions, with the free spectral range constrained only by the span of the combs.



a Femtosecond laser



b Electro-optic comb



c Kerr comb generation

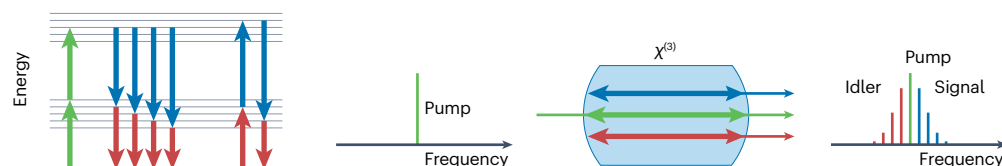


Fig. 3 | Common frequency-comb generators for dual-comb spectroscopy. **a**, Short laser pulses circulate in a resonator that contains a pumped broad-bandwidth laser gain medium and a saturable absorber (SA). The spectrum of the out-coupled train of pulses forms a frequency comb. **b**, By modulating the phase and the amplitude of a continuous-wave electric field in a medium with second-order nonlinearity ($\chi^{(2)}$) driven by a radio-frequency generator, side bands are generated, evenly spaced at the driving frequency. If no resonator is used, the phase noise of the radio-frequency source multiplies with distance from the carrier. **c**, A low-loss resonator with third-order nonlinearities ($\chi^{(3)}$) is excited by a continuous-wave laser. By cascaded four-wave mixing, phase-locked modes are generated over a broad span about the continuous-wave laser frequency. Panels **a** and **c** adapted from ref. 185, Springer Nature Limited.

generators is therefore particularly attractive for a portable, battery-operated dual-comb spectrometer. However, achieving mode-locking in such devices may be challenging owing to intracavity phase distortions. Nevertheless, several semiconductor-based comb generators have been reported^{90–93}, including frequency-modulated combs which do not emit pulses^{94–97} and optically pumped external-cavity semiconductor disk lasers^{62,98}. The possibility of designing the emission range is another attractive feature. Quantum cascade lasers and interband cascade lasers directly emit in the mid-infrared spectral region, which makes them appealing for molecular fingerprinting^{99–101}. Challenges include their rather narrow span, their repetition frequency of typically 10 GHz, which is often too high for gas sensing without interleaving, and the absence of any solutions to direct referencing to a radio-frequency clock. Mode-locked lasers on silicon or silicon nitride waveguides can show a remarkably small repetition frequency for chip-scale devices¹⁰², which makes them suited for gas-phase dual-comb spectroscopy, although their emission is currently limited to the near-infrared region⁹¹.

Kerr combs. A Kerr comb (Fig. 3c) is a comb generated by parametric oscillation in a resonator injected by a continuous wave pump laser¹⁰³. Typically, the resonator is a microresonator on a chip, which features a high Kerr nonlinearity, a high Q factor and a diameter from micrometre to millimetre. With continued progress to optical integration⁶¹, such microcombs hold much promise for miniaturized dual-comb spectrometers and provide access to a parameter range inaccessible to most other frequency-comb generators: their line spacing typically lies in the range from 10 GHz to 1 THz. Therefore, Kerr combs present an untapped potential for dual-comb spectroscopy^{104–106}, especially in condensed matter^{107,108} and for hyperspectral holography¹⁰⁹. The first proof-of-principle demonstrations have, however, uncovered some of the challenges related to the requirement for a high

dynamic range and the lack of convenient sources outside the telecommunication region¹⁰⁸.

Nonlinear frequency conversion of frequency combs.

Frequency-comb technology is most advanced in the near-infrared spectral region. However, for spectroscopy, the mid-infrared, terahertz and UV regions are more commonly studied, as they include intense signatures of a high number of atoms and molecules. An effective approach for reaching these regions has been nonlinear frequency conversion (Fig. 4). Nonlinear processes such as difference frequency generation¹¹⁰, optical parametric oscillation¹¹¹, four-wave mixing¹¹² or harmonic generation³² has proven effective for reaching relevant regions (Fig. 5). For this purpose, a variety of crystals, fibres and platforms for nanophotonic waveguides on photonic chips have been successfully used in conjunction with frequency-comb generators¹¹³. Nevertheless, efficient conversion at low pulse energy, as encountered with systems of high repetition frequency, remains a challenge. Powerful near-infrared frequency-comb generators combined with lithium niobate technology enable efficient frequency-comb generation to wavelengths as long as 5 μm (refs. 110,114–116). Although impressive demonstrations have been published recently at long infrared wavelengths^{117,118} or in the ultraviolet region at wavelengths shorter than 400 nm (refs. 119–122), dual-comb spectroscopy in these regions remains challenging and would benefit from further advances, particularly in light-source design. A recent notable study highlights the potential of high-harmonic generation in a rare gas to enable dual-comb spectroscopy at 149 nm (ref. 123).

Results

This section covers the key aspects of computing and evaluating spectra in dual-comb spectroscopy, highlighting the techniques, challenges and considerations involved in obtaining accurate and reliable spectral data.

Spectrum computation

We describe the steps involved for revealing the spectrum from the interferogram using the Fourier transformation as a harmonic tool. This calculation procedure is independent of the sample location: it can be placed on the beam of one comb (dispersive configuration) or after the two comb beams have been combined. When comb line resolution is not sought for, processing an interferogram is identical to that in Fourier-transform spectroscopy. Thus, textbooks on Fourier-transform spectroscopy provide a valuable introduction to calculating spectra¹²⁴.

The step-by-step procedure illustrated in Fig. 6 focuses on calculating a spectrum for reaching comb line resolution, although it remains fully valid at lower resolutions. A double-sided interferogram is extracted from the time-domain recording (Fig. 6a). A double-sided interferogram samples the interference over optical delays between $-n/(2f_{\text{rep}})$ to $+n/(2f_{\text{rep}})$, where n is an odd integer, $n=1$ being sufficient. $n=1$ corresponds to the comb line resolution where all the spectroscopic information is revealed. If $\Delta f_0 = 0$ (modulo Δf_{rep}), and the digitization rate is an integer multiple of the repetition-rate difference Δf_{rep} , then extending the recorded time sequence and Fourier transforming it does not reveal any additional information. However, resolving each comb line is useful when the comb and sampling conditions cannot be adjusted or when the instrumental line shape is being scrutinized. Zero-filling can be performed symmetrically at the two ends of the interferogram (Fig. 6b). Filling with six times more zero than there

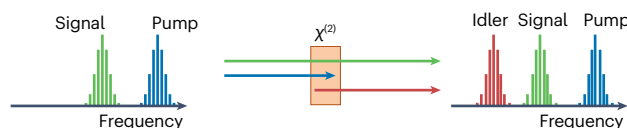
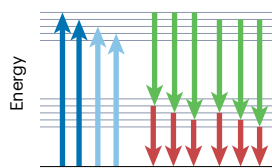
are samples usually leads to good results. The added zeros interpolate the spectrum but evidently do not improve the resolution. The time-domain data, $I(t)$, are shifted about the intensity maximum to flatten the phase slope and reference it to the zero phase (Fig. 6c). Using a complex Fourier transformation (FT) with real (Re) and imaginary (Im) parts (Fig. 6d), the complex spectrum, B_{complex} , as a function of frequency, ν , is

$$B_{\text{complex}}(\nu) = \text{Re}(\text{FT}(I))(\nu) + i \text{Im}(\text{FT}(I))(\nu), \quad (2)$$

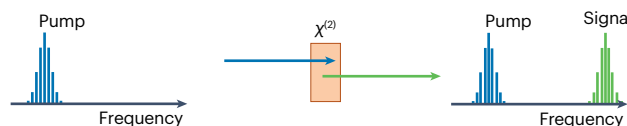
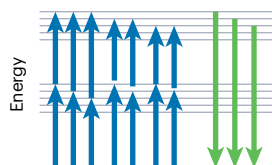
where i is the imaginary unit. The phase, ψ , and the amplitude, B , can be extracted from B_{complex} .

The phase spectrum is $\psi(\nu) = \arctan(\text{Im}(\text{FT}(I))(\nu)/\text{Re}(\text{FT}(I))(\nu))$. The phase ψ is not defined when the real part of the complex transform $\text{Re}(\text{FT}(I))$ is equal to zero. Therefore, the phase spectrum ψ should be computed only at the comb line positions (Fig. 6e) and interpolated in between (Fig. 6f). The phase spectrum contains the signature of all the asymmetries in the interferogram. There are multiple reasons for such asymmetries in a dual-comb spectrometer. For example, the position of zero optical delay does not usually coincide with a sample at which the interferogram is digitized. Optical delay can be frequency-dependent owing to dispersion in the instrument, arising from chirped lasers, unbalanced optical elements or frequency-dependent electronic response. This necessitates considering a symmetric range of optical

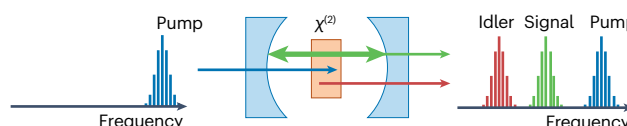
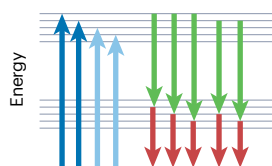
a Difference frequency generation



b Sum frequency generation



c Optical parametric oscillation



d Four-wave mixing

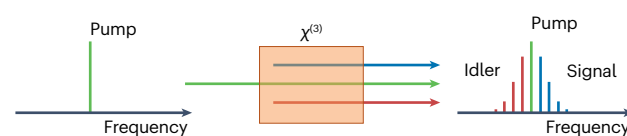
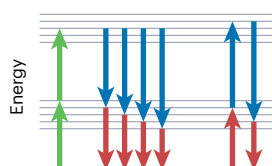


Fig. 4 | Spectral broadening and nonlinear frequency conversion of frequency combs. a, Difference frequency generation in which two waves mix in a medium with second-order nonlinearity ($\chi^{(2)}$), generating a third wave with a frequency equal to the difference between the frequencies of the incident photons. The many modes of either the signal or the pump combs (or both) lead to a broad idler comb of many difference frequencies. **b**, Sum frequency generation in which two waves can mix in a medium with second-order nonlinearity, generating a third wave with a frequency equal to the sum between the frequencies of the incident photons. **c**, Optical parametric oscillation in which an optical resonator contains a medium with second-order nonlinearity is optically pumped, provided coherent oscillation of the signal comb and of the idler comb. **d**, Four-wave mixing in which three frequencies create a fourth frequency in a highly confined medium with third-order nonlinearities ($\chi^{(3)}$), such as a waveguide or an optical fibre. Panels **a** and **c** adapted from ref. 185, Springer Nature Limited.

Table 2 | Comparison of the main types of frequency-comb generators

Attribute	Fibre laser	Electro-optic comb	Kerr comb	Semiconductor laser	Solid-state laser oscillator
Repetition frequency	80×10^6 to 250×10^6 Hz	1×10^3 to 30×10^3 Hz	2×10^9 to 1×10^{12} Hz	1×10^9 to 25×10^9 Hz	80×10^6 to 10×10^9 Hz
Centre frequency (wavelength)	192 THz (1,560 nm), 288 THz (1,040 nm)	192 THz (1,560 nm)	192 THz (1,560 nm)	37 THz (8.1 μ m) for quantum cascade laser, 90 THz (3.2 μ m) for interband cascade laser, 190 THz (1.5 μ m for mode-locked laser on Si or SiN)	291 THz (1,030 nm), 193 THz (1,550 nm), 125 THz (2,400 nm)
Average power	>50 mW oscillator >250 mW with Er amplifier >10 W with Yb amplifier	>1 mW	<1 mW for bright soliton microcomb	>1 W for quantum cascade laser	60 mW (ref. 180), >1 W for Ti:Sa (ref. 169), >15 W for thin-disk ⁷¹
Integrated phase noise of a comb line	<55 mrad (1 Hz to 1 MHz) ¹⁸¹ Out of loop	Increases linearly with mode numbers. Mostly influenced by the radio-frequency synthesizer, which drives the phase modulators ⁸⁴	Not thoroughly studied in the literature	200 mrad (1 Hz to 1 MHz) ¹⁸² In loop, with optical injection locking	40 mrad (100 Hz to 10 MHz) In loop ¹⁸⁰
Intensity noise	<0.005% (1 Hz to 1 MHz) for $f_{\text{rep}} = 250$ MHz (ref. 77)	Not thoroughly studied in the literature	0.034% (100 Hz to 10 MHz) for $f_{\text{rep}} = 89$ GHz (ref. 183)	Not thoroughly studied in the literature	<0.01% (1 Hz to 1 MHz) for $f_{\text{rep}} = 1$ GHz (ref. 180)
Bandwidth of actuators	Two fast actuators >1 MHz (ref. 79)	Two fast actuators >100 kHz (ref. 38)	Two actuators >100 kHz (ref. 184)	At best two actuators >30 kHz (refs. 182,184)	At best two actuators <50 kHz (ref. 180)
9Span	>10 THz at oscillator output	<100 $\times f_{\text{rep}}$	<20 THz (ref. 107)	<1 THz	>10 THz at oscillator output
Number of comb lines	>10 ⁵ at oscillator output >10 ⁶ with spectral broadening	<100	<100	<100	>10 ⁵ at oscillator output >10 ⁶ with spectral broadening
Full stabilization against radio-frequency clock	Yes	Not possible in standard configurations	Not possible in standard configurations	Not possible in standard configurations	Yes
Full stabilization against hertz linewidth optical reference	Yes	Not possible in most configurations	Possible but not implemented	Not possible in most configurations	Possible but not implemented
Tunability of f_{rep}	1 MHz for 100-MHz rep frequency	Can be over several gigahertz	Limited	Limited	10 kHz for 100-MHz f_{rep}
Fine tuning of f_0	Continuously tunable over (0, f_{rep})	Continuously tunable over (0, f_{rep})	Limited	Limited	Limited
Footprint	1 m ²	0.1 m ²	1 m ²	0.1 m ²	1 m ²

The numbers given illustrate performance reported in dual-comb spectroscopy experiments but do not cover all specific implementations in dual-comb spectroscopy nor applications other than dual-comb spectroscopy. f_{rep} is the repetition frequency.

delays. In addition, if only one comb interacts with the sample, the phase spectrum contains the dispersion caused by the sample.

The amplitude spectrum, B , is a spectrum of real values (not a complex number), which provides the true representation of the spectrum (Fig. 6g), defined by

$$B(\nu) = \text{Re}(\text{FT}(I))(\nu) \cos(\psi(\nu)) + \text{Im}(\text{FT}(I))(\nu) \sin(\psi(\nu)). \quad (3)$$

The amplitude spectrum can take positive and negative values, as the noise and the instrumental line shape can generate negative samples. In any Fourier-transform spectroscopy technique, the spectrum shows the optical power encoded into an amplitude representation. Using double-sided interferograms strongly benefits the phase determination, whose baseline can be interpolated and extrapolated by a fit to a polynomial of the lowest possible order¹²⁵. Computing the amplitude corrected by the phase function (calculated over the same optical-delay span as the amplitude) ensures that the Fourier-transformed spectrum accurately represents the true spectral

content of the sample. This step is crucial for achieving high-resolution spectra, as any uncorrected phase variations can lead to distortions and inaccuracies in the spectral data. The amplitude spectrum B and the phase spectrum ψ can be processed to lead to a transmittance spectrum (Fig. 6h) and a dispersion spectrum (Fig. 6i). Computing the magnitude spectrum $B_{\text{mag}} = ([\text{Re}(\text{FT}(I))(\nu)]^2 + [\text{Im}(\text{FT}(I))(\nu)]^2)^{1/2}$ (or the power spectrum B_{mag}^2) from a double-sided interferogram also drops out the phase function, but it only provides the modulus of the spectrum. The noise becomes nonlinear and does not average out as efficiently as it could. With an increasing number of averaging, it creates a background of increasing magnitude that can limit the contrast of the comb lines; this procedure should be avoided as much as possible.

Averaging interferograms in the time domain or spectra in the frequency domain is often necessary to enhance the signal-to-noise ratio and to improve the precision of the spectral data. Although time-domain averaging reduces computational time, it requires identical interferograms — a condition met when $\Delta f_0 = 0$ (modulo Δf_{rep}) and the digitization rate is an integer multiple of Δf_{rep} . Frequency-domain

averaging involves averaging the amplitude spectra, each individually corrected from the phase. Computation time is substantially longer because a Fourier transform must be computed for each averaging step. However, this approach accounts for individual phase functions, which is essential when the interferometer lacks sufficient phase stability for long-term time-domain averaging. Mixed solutions where series of individual interferograms are averaged over a few seconds and then Fourier transformed and further averaged in the frequency domain offer realistic compromises between complex instrumentation and performance.

Digital corrections

Besides asymmetries in the interferogram caused by imperfections in the optical and electronic layout, the interferogram in dual-comb spectroscopy may also be sampled at uneven effective optical delays owing to relative timing and phase fluctuations between the combs. Uniform time sampling does not necessarily correspond to uniform optical-delay sampling. When this correspondence cannot be maintained experimentally, it can be reconstructed a posteriori by digitally correcting both the time axis and the interferometric phase evolution. Although this procedure is often referred to as phase correction, it differs fundamentally from classical Fourier-transform phase correction, as it involves reconstruction of the delay coordinate itself. For clarity, we therefore refer to this process as digital correction of the interferogram.

Digital correction is independent of the location of the sample, which can be placed on the beam of one comb (dispersive configuration) or after the two comb beams have been combined. Digital correction of the interferogram consists of tracking the relative variations of the combs for correcting the interferogram digitally or in applying

optimization procedures based on assumptions on the expected interferogram. Sophisticated tracking experimentally monitors the variations of the two beat notes between pairs of interfering comb lines and uses these signals to a posteriori resample the interferogram and construct a phase-stable one³³. More recent techniques without any tracking rely on the assumption that the dual-comb signal should be composed of a sum of signals at discrete radio frequencies and applies an optimization procedure for correcting the interferogram, which works best when the fluctuations remain smaller than $\Delta f_{\text{rep}}/2$ (refs. 35,126). Several algorithms can improve the spectra, with features that make them better suited to particular dual-comb generators^{34,127,128}. Software corrections can facilitate the shifting of the phase in the interferograms, enabling real-time averaging and the use of comb generators where the parameters are not fully controllable. These digital corrections can be applied in real time, using field-programmable gate arrays³³ or graphics processing units^{129,130}, which is useful for reducing data storage requirements. It remains important to carefully validate the phase-correction process for any signs of residual errors or artefacts in the spectrum, as digital corrections can potentially introduce artefacts or distortions if the correction is not properly applied or if the relative fluctuations are too large. Moreover, these techniques may be less effective when the signal-to-noise ratio is poor or under photon-counting conditions^{131,132}. The impact of the various correction techniques on the overall accuracy and reliability of the spectral data needs further investigation. Careful validation by comparing precise data against state-of-the-art molecular line parameters measured by other techniques has been realized in several occurrences^{30,133,134}. However, as dual-comb spectroscopy should aim beyond the state of the art, careful evaluation of all systematic effects including those generated by

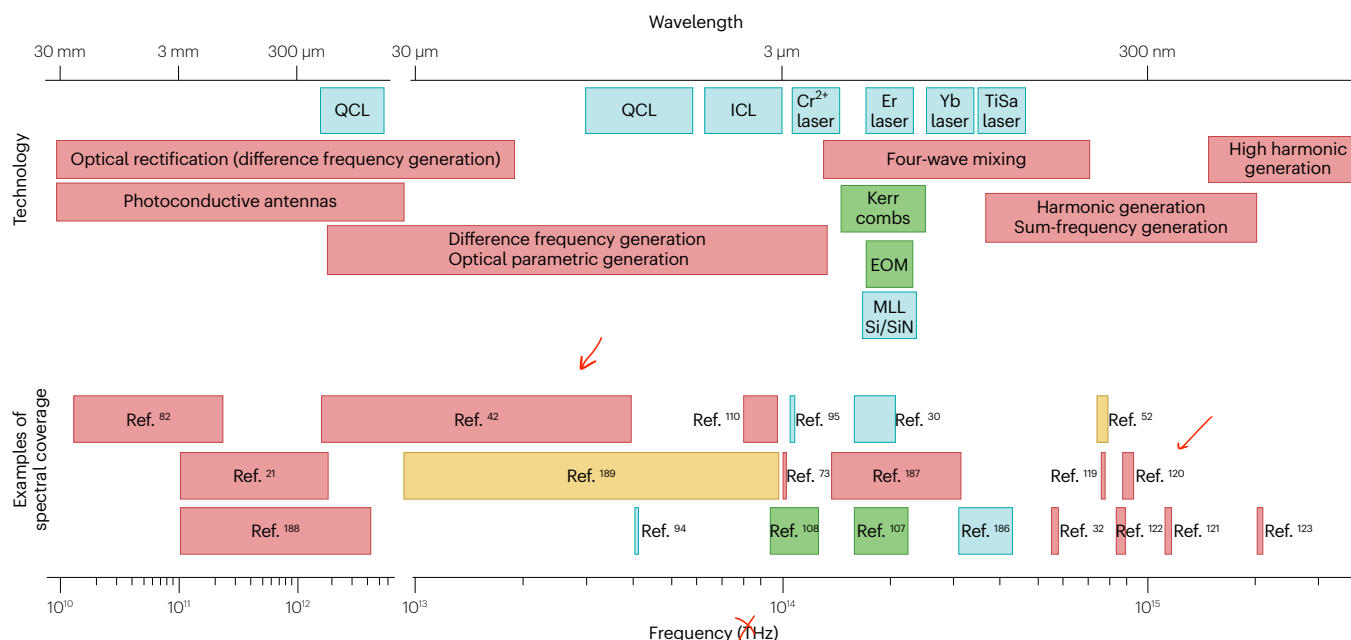
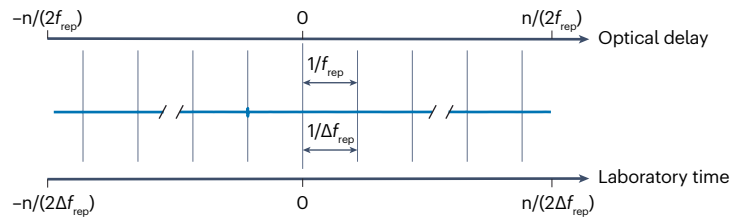


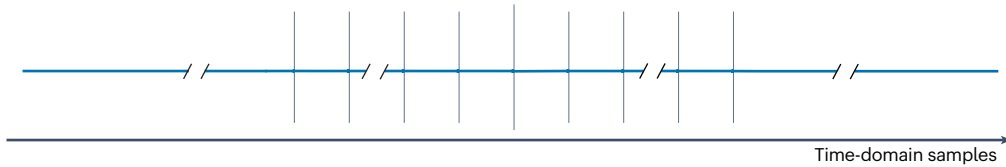
Fig. 5 | Main technologies for frequency-comb synthesizers and spectral coverage of dual-comb spectroscopy. Overview of the laser technologies available for dual-comb spectroscopy as a function of spectral range. The radiation from self-oscillating laser systems (blue)^{30,94,95,186} and driven combs (green)^{107,108} can be converted to other ranges using nonlinear techniques (pink)^{21,32,42,73,82,110,119–123,187,188}. The spectral span obtained in some selected

dual-comb linear and nonlinear (yellow)^{52,189} spectra is provided for illustrative purposes and does not provide an exhaustive report of all the capabilities of dual-comb systems. Please note that the x scale differs over the span of 10^{10} to 10^{13} THz and at frequencies higher than 10^{13} THz. EOM, electro-optic modulators; ICL, interband cascade laser; MLL, mode-locked laser; QCL, quantum cascade laser.

a Double-sided interferogram I



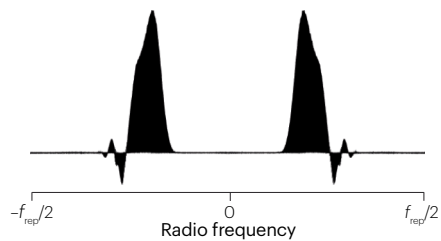
b Interferogram I with zero filling



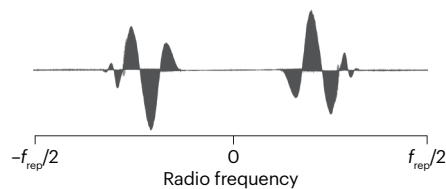
c Shifted interferogram I



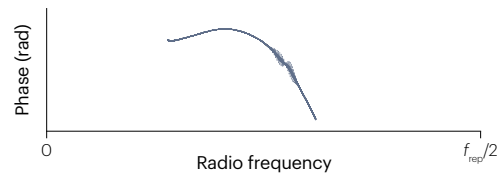
d Real part of the Fourier-transform $\text{Re}(\text{FT}(I))$



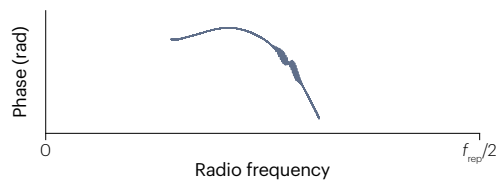
Imaginary part of the Fourier transform $\text{Im}(\text{FT}(I))$



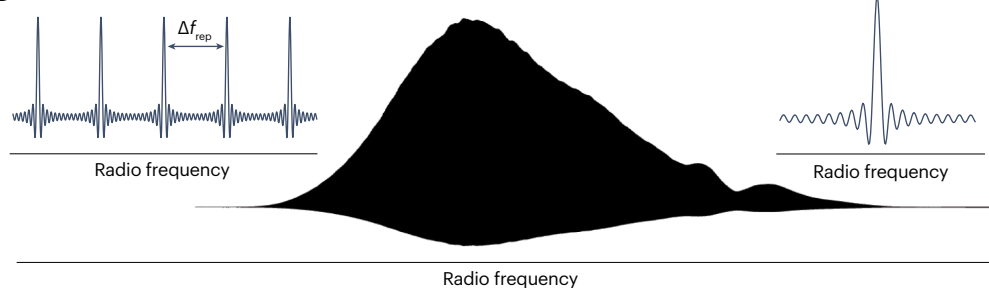
e Phase spectrum $\arctan(\text{Im}(\text{FT}(I))/\text{Re}(\text{FT}(I)))$



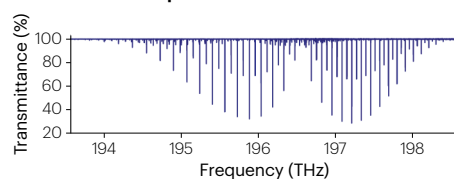
f Interpolated/extrapolated phase spectrum



g Amplitude spectrum B



h Transmittance spectrum



i Dispersion spectrum

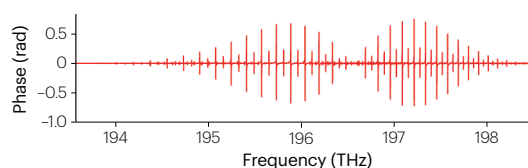


Fig. 6 | Computing a spectrum from an interferogram. A double-sided interferogram, $I(t)$, is acquired (panel **a**). For interpolation in the spectrum, zeros are added on both sides of the interferogram (panel **b**). The interferogram is shifted to minimize the slope of the phase spectrum, $\psi(v)$, where v is the frequency (panel **c**). The complex Fourier transform, B_{complex} , of the interferogram is calculated, leading to the real, $\text{Re}(\text{FT}(I))(v)$, and imaginary, $\text{Im}(\text{FT}(I))(v)$, parts of the spectrum (panel **d**). In the regions where the signal is strong only, such

as in the case of maxima of comb lines of good signal-to-noise ratio, the phase spectrum is calculated (panel **e**). The phase spectrum is interpolated and can be extrapolated, if needed (panel **f**). The interpolated phase spectrum enables calculation of the amplitude spectrum, $B(v) = \text{Re}(\text{FT}(I))(v) \cos(\psi(v)) + \text{Im}(\text{FT}(I))(v) \sin(\psi(v))$, which is the true representation of the spectrum (panel **g**). Further processing of the baseline leads to the transmittance spectrum (from B) (panel **h**) and the dispersion spectrum (from ψ) (panel **i**). f_{rep} , repetition frequency.

digital correction techniques still require deeper investigation. These digital correction techniques do not address the absolute stability of the combs, for example if they are kept free-running.

Evaluating a spectrum

The evaluation of a spectrum obtained by dual-comb spectroscopy encompasses both the assessment of instrumental performance and the analysis of spectroscopic data, which is often the primary scientific objective. This evaluation requires appropriate representation of the spectrum, a quantitative understanding of noise sources and careful assessment of statistical and systematic uncertainties. Showing a spectrum with resolved, narrow comb lines – now a hallmark of many dual-comb experiments – provides a particularly direct means of assessing instrumental performance, as it enables straightforward inspection of the instrumental line shape.

The instrumental line shape is a critical factor affecting spectral resolution and precision. In general, it can be defined as the instrument's spectral transfer function, describing how an infinitesimally narrow spectral feature is mapped onto the measured spectrum by the spectrometer's resolution, sampling and coherence properties. With a dual-comb interferometer, resolving the individual comb lines provides direct access to the instrumental line shape for each line³⁰. In an amplitude spectrum with resolved comb lines, the instrumental line shape is expected to be a cardinal sine function, which narrows with increasing measurement time because of finite time-window apodization. Appropriate sampling conditions prevent unwanted interference between the sidelobes of neighbouring comb lines, ensuring that each line remains unaltered by the instrumental line shape of adjacent lines. Alternatively, such effects can be minimized by transforming long interferograms containing many recurrences, for which the ringing of the cardinal sine becomes small compared with the comb line spacing.

A central parameter in spectral evaluation is the coherence time, defined as the duration over which the relative phase between the two combs remains stable. The coherence time constrains the achievable spectral resolution, accuracy and signal-to-noise ratio, and limits the extent to which interferograms can be coherently averaged. In practice, it is useful to distinguish between the coherence time and the maximum averaging time over which the signal-to-noise ratio continues to improve with the square root of time, as the latter may be longer and depends on both the instrument and the data-processing strategy.

Statistical uncertainty in the spectrum arises from random noise sources, including thermal noise, electronic noise, laser intensity noise and phase noise associated with the combs and interferometric detection. The signal-to-noise ratio provides a convenient measure of spectral quality, but its numerical value depends critically on how it is defined. Noise reduction techniques such as balanced or differential detection, coherent averaging, filtering can improve spectral quality. However, the signal-to-noise ratio often reaches a saturation limit imposed by systematic effects. Systematic effects, including instrumental drifts, environmental perturbations, parasitic

etalon effects, residual phase errors and sample-related artefacts, frequently dominate the limitations on accuracy and long-term stability. These effects can distort line shapes, shift line centres and bias intensity measurements, and therefore require careful calibration, experimental control and, where appropriate, correction during data processing.

Spectral resolution in dual-comb spectroscopy is a subtle concept. In the case of free-running combs, experimental characterization of the comb instabilities under the conditions of measurement is often necessary. Phase correction is another critical component of spectral evaluation, and the applied correction procedures and their potential impact on systematic errors must be carefully assessed.

Extracting spectral line parameters – such as line positions, widths and intensities – or derived physical quantities such as concentration or temperature is a central step in data analysis and is frequently the final scientific goal. The achievable precision and accuracy depend on the stability of the experimental set-up, the quality of the interferograms, the instrumental line shape and the treatment of noise and systematic effects. Comparison with reference data obtained using state-of-the-art techniques is often essential for validating the results.

Applications

This section outlines the unique features and applications of dual-comb spectroscopy across various fields, highlighting its potential to address critical scientific questions and advance our understanding of complex systems. Dual-comb spectroscopy is highly versatile and can be combined with other powerful instrumentation and sampling techniques, for additional diagnostics and capabilities (Box 3).

Laboratory gas-phase spectroscopy

Determination of line parameters. Much of the scientific literature on dual-comb spectroscopy is still largely focused on describing new experimental schemes, and there are relatively few spectroscopic studies for laboratory spectroscopy. However, those that do exist are highly instructive about the performance of dual-comb spectroscopy and are expected to inspire further research within the growing community (Fig. 7).

Most spectroscopic work so far has been performed for linear absorption in the infrared range. Because the line spacing of conveniently-available frequency combs is in the range 80–250 MHz, dual-comb spectroscopy has been directly suited to the interrogation of small molecules in the gas phase, in conditions where the Doppler and collisional effects dominate the spectral line shape. In dual-comb spectroscopy, the unique combination of high resolution, broad spectral bandwidth and accuracy makes it a powerful tool for investigating spectral profiles and understanding molecular structure and dynamics.

The first key advantage of dual-comb spectroscopy for line position measurement is the direct calibration of the frequency scale directly to an atomic clock. This strongly contrasts with spectrometers which do not use a comb line source or do not resolve its lines

Box 3 | Sampling techniques associated with dual-comb spectroscopy

Fourier spectroscopy techniques are highly versatile and can be efficiently integrated with various sampling methods and instrumentation to enhance analytical capabilities across multiple scientific disciplines. For a better sensitivity to weak absorptions, cavity enhancement can increase the absorption path^{20,75,196–198} or surface enhanced absorption can lead to electromagnetic field enhancement when the molecules are placed at the interface with a metal or a metal oxide¹⁹⁹. Hyperspectral imaging can be implemented by replacing a single photodetector with a matrix of detectors, enabling the simultaneous measurement of as many spectra as there are detector pixels^{174,200–203}. With a suitable optical layout and data processing, hyperspectral 3D imaging can be implemented in the form of digital holography, leading to one hologram per comb line²⁴. Other schemes practice frequency to space conversion to associate one comb line to one spatial location^{204,205}, trading the spectroscopic capabilities to the imaging ones. With a single photodetector, raster scanning the beam relative to the sample enables hyperspectral mapping^{46,160}, near-field microscopy²⁰⁶ and microscopy^{207,208}, whose sensitivity can be enhanced by detection schemes such as photothermal detection¹⁶¹ or cavity enhancement²⁰⁹. Time-resolved

detection facilitates the monitoring of transient phenomena^{39,139,140,142}. In the mid-infrared region, up-conversion spectroscopy^{40,43}, including electro-optic sampling^{41,42}, provides alternative ways to Raman spectroscopy^{45–47,49,169} to interrogate fundamental vibrations while detecting in the near-infrared or visible regions where detectors are more sensitive. Stepping mode operation decorrelates experimental time from interferometric delay scanning, providing flexibility in new modes of operation^{174,210}. Photon counting^{119,132,211} opens up interesting prospects for spectroscopy at very low light levels²¹¹ or ghost imaging²¹², whereas the use of squeezed states of light or of quantum correlations enables signal-to-noise ratios beyond the shot-noise limit^{191,213}. Evanescent sensing in the gas phase enables interrogation of very small volumes of gas²¹⁴. Background-free detection techniques such as fluorescence⁵¹, photoacoustic^{215,216} or photothermal detection¹⁶¹ also enhance the detection sensitivity. Polarization sensitive dual-comb spectroscopy, such as dual-comb ellipsometry²¹⁷ or optical activity spectroscopy²¹⁸, reveals specific properties in thin films or chiral molecules. Arbitrary light sources, including incoherent ones, can be investigated through their beating with two combs²¹⁹.

where it is necessary to calibrate the wavenumber scale of the spectrum against reference spectral lines. The comb lines offer calibration markers across the entire span, whereas the use of reference spectral lines (independently of their accuracy) has shown to be prone to systematic effects, particularly when the references are not spread across the spectrum. When combined with the improved signal-to-noise ratio over techniques using incoherent light sources, dual-comb spectrometers achieve roughly an order-of-magnitude improvement in line position accuracy compared with Fourier-transform spectrometers with incoherent light sources. Reported uncertainties range from hundreds of kilohertz to a few megahertz^{30,133,134}. Measurements of line positions in dual-comb spectroscopy are often limited by statistical uncertainty: the precision with which the line centre of a profile can be determined is proportional to the ratio of the profile linewidth to the signal-to-noise ratio, which leads to the quoted uncertainty range. Dual-comb spectroscopy has therefore not yet reached its full potential for line-position measurements. Accessing narrower spectral profiles – for example from cold molecules or from Doppler-free schemes – holds great promise for unlocking this potential.

Another crucial feature in dual-comb spectroscopy is the narrow and well-defined instrumental line shape. The width of the optical comb lines of a comb stabilized against a radio-frequency clock typically have a width on the order of 100 kHz at 1 s; this width can be much narrower if the comb is stabilized to an optical reference². A width of 100 kHz is negligible compared with the Doppler or collisional width of small molecules in the gas phase in the optical domain. For example, the Doppler width of CH₃I at a centre frequency of 30 THz (10 μ m) is 30 MHz at room temperature – larger than the width of the lines of any stabilized comb. Dual-comb spectrometers are thus well suited to the study of line parameters beyond the line positions, such as line intensities and line shapes. Line shape analysis provides detailed information about intermolecular interactions, environmental conditions and dynamic processes and thus is key to a better understanding of collisional

processes, to testing potential energy surfaces and for extracting gas media properties from sounding techniques¹³⁵. Dual-comb set-ups for line shape studies often involve using high-pressure gas cells and high temperatures and derive collisional parameters beyond the Voigt profile^{136,137}.

Doppler-free spectroscopy. The potential of dual-comb spectroscopy for high accuracy over a broad spectral bandwidth has not yet been realized, as the precision has mostly been limited by statistical uncertainty in measuring Doppler-broadened transitions. Doppler-free spectroscopy is essential for high-precision measurements¹³⁸. The combination of dual-comb spectroscopy with nonlinear spectroscopic schemes cancels Doppler broadening, as demonstrated with Doppler-free dual-comb two-photon excitation^{50,52}. Excitation by the two counter-propagating comb beams, which form two standing-wave trains of pulses with slightly different repetition frequencies, cancels the Doppler effect to a large extent. In the first demonstrations^{50,52}, the resolution was improved by spectral interleaving, which is a lengthy process¹⁸. Another step towards broadband frequency metrology has been obtained with the recent demonstration of a dual-comb interferometer based on self-referenced fibre lasers, capable of recording spectra spanning over 1 THz with a line spacing of 1 MHz (ref. 26). This results in resolving powers higher than 2×10^8 obtained in a single measurement, without the need for interleaving²⁶. Further development of dual-comb Doppler-free techniques and their extension to new spectral regions can address issues such as the determination of fundamental physical constants, the testing of ab initio quantum chemical chemistry and the refinement of molecular potential energy surfaces.

Transient species. The ability to study transient species, such as short-lived intermediates and radicals, is crucial for understanding chemical reaction kinetics and atmospheric processes. The synergy of rapid measurement times and often moderately broad

spectral bandwidths enhances time-resolved spectroscopy across various applications^{39,139–143}. In most demonstrations, a synchronization between the transient phenomenon and the data acquisition has been implemented, followed by measuring of an interferogram every $1/\Delta f_{\text{rep}}$ over a time which can be shorter than $1/\Delta f_{\text{rep}}$. The time resolution, given by $1/\Delta f_{\text{rep}}$, can be higher if combs of large repetition frequencies¹⁴⁰ or of narrow spans^{139,143} are used. Although, in

principle, the acquisition scheme is suited to non-repetitive phenomena, the need for synchronization and high signal-to-noise ratio have favoured the study of repetitive, triggerable phenomena. Key challenges include enhancing the detection sensitivity to capture weak transient signals and improving the temporal resolution while preserving high resolution and spectral bandwidth to resolve rapid dynamic processes.

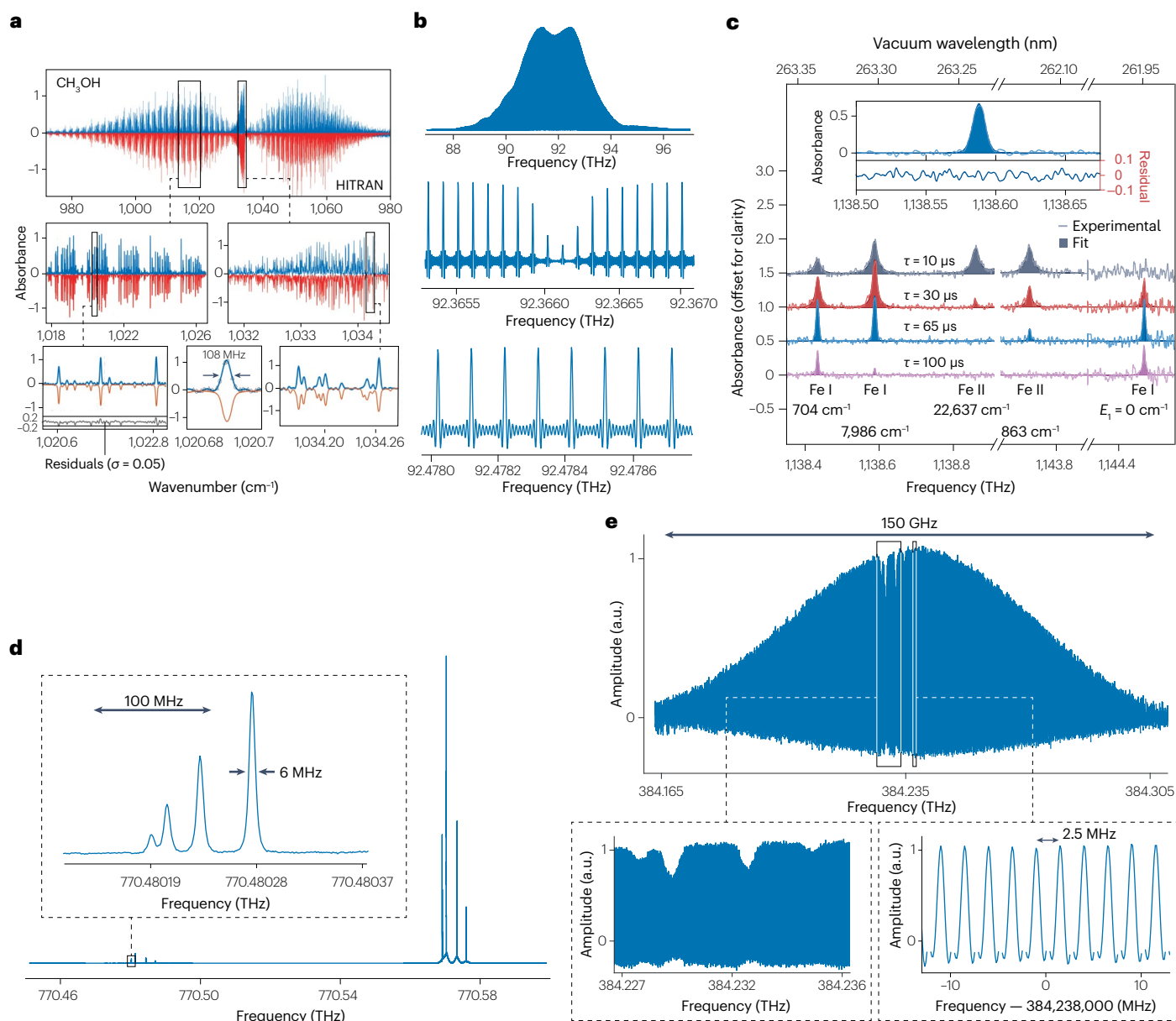


Fig. 7 | Selection of experimental dual-comb spectra. a, Mid-infrared absorbance spectrum of methanol at $10\ \mu\text{m}$ with an acquisition time of 12 s. **b**, Mid-infrared spectrum of ethylene with 82,000 resolved comb lines spaced by 100 MHz, recorded within 1,742 s in the $3\text{-}\mu\text{m}$ region. **c**, Deep ultraviolet absorbance spectra at a resolution of 1.2 GHz after laser ablation of an iron metal sample. Transitions in Fe I and Fe II are observed. **d**, Near-ultraviolet Doppler-free two-photon excitation spectrum of the $5\text{S}-5\text{D}$ transitions in Rb measured with a resolution of 1 MHz obtained by interleaving 100 spectra

at 100 MHz comb line spacing. **e**, The near-infrared spectrum of the $5\text{S}-5\text{P}$ transitions in Rb is measured with a comb of a line spacing of 2.5 MHz. With a traditional spectrometer, a resolution of 2.5 MHz would require a path difference excursion of 100 m. a.u., arbitrary unit. Panel **a** reprinted with permission from ref. 117, Optica. Panel **b** reprinted with permission from ref. 31, PNAS. Panel **c** adapted with permission from ref. 121, Optica. Panel **d** adapted with permission from ref. 52, Optica. Panel **e** adapted with permission from ref. 26, Optica.

Sensing

Environmental sensing. Dual-comb spectroscopy is improving environmental sensing by enabling the precise and real-time monitoring of atmospheric gases, pollutants and greenhouse gas concentrations. The technique's ability to provide high-resolution spectra over broad spectral ranges enables the detection and quantification of multiple species simultaneously, which is crucial for assessing air quality and tracking emissions. For environmental applications, open-path dual-comb spectroscopy samples air over long distances (hundreds of kilometres)^{144–148}. Results include tracking green-house gases in urban¹⁴⁹, agricultural¹⁵⁰ and gas production¹⁵¹ locations, volatile organic trace compounds^{146,152} or air pollutants formed during the combustion of fossil fuels¹⁵³ and address questions related to the sources, sinks and kinetics of atmospheric constituents and their impact on climate and human health. Recent advances include the development of portable dual-comb spectroscopy systems for field measurements and progress towards integration with remote sensing platforms for wide-area monitoring¹⁵¹. Challenges include dealing with varying and potentially extreme environmental conditions, reaching the necessary sensitivity for detecting small concentrations within a short time and ensuring the robustness of the systems for long-term deployments of remote sensing platforms onboard balloons, planes or satellites.

Plasma diagnostics. In plasma diagnostics, dual-comb spectroscopy offers a non-intrusive method for studying the complex chemical processes and reactions that occur in laser-induced plasmas or in flames and engines^{154,155}. Its good temporal and spectral resolution make it ideal for analysing the formation and breakdown of intermediates and pollutants¹⁵⁶. Plasma diagnostics using dual-comb spectroscopy often involve probing hot gases with high-speed dual-comb systems to capture rapid changes in chemical composition and temperature. Results include detailed spectra of plasma species, providing insights into reaction pathways and energy release mechanisms^{157,158}. Key questions include understanding the mechanisms of fuel oxidation, pollutant formation and the efficiency of combustion processes. The main challenges involve maintaining high measurement sensitivity in high-temperature, turbulent environments and developing robust systems capable of operating in such extreme conditions.

Condensed phase

It may sound counter-intuitive to use frequency combs for interrogating condensed matter as the transitions in the condensed phase are often very broad and thus do not require a high precision, but a number of proof-of-principle demonstrations of measurement techniques have highlighted interesting potential for investigating molecular vibrations and interactions in complex media^{45,46,159}. Linear hyperspectral mapping^{46,160,161} and imaging^{24,109}, nonlinear dual-comb spectroscopy^{45,46}, subsequent asynchronous pump-probe¹⁶² and multidimensional comb spectroscopy have been mostly demonstrated in the condensed phase. Although not involving combs, asynchronous sampling with mode-locked lasers provides unique insights into electronic relaxation processes and solvent dynamics by permitting the simultaneous monitoring of multiple electronic states and their interactions with the solvent environment^{57,163}. This capability is essential for understanding energy transfer processes in chemical and biological systems. For these studies, dual-comb spectroscopy set-ups often include time-resolved measurements to capture the dynamics of electronic relaxation and solvent interactions¹⁶⁴. Results typically feature spectra that reveal the timescales and pathways of energy dissipation

and redistribution. Key challenges involve achieving the complexity of schemes of multidimensional spectroscopy^{58,165}, the temporal resolution needed to capture fast relaxation processes and developing techniques to separate electronic and solvent contributions to the spectra. The need for high sensitivity and resolution to detect subtle vibrational features and the complexity of analysing coupled electronic and vibrational systems in biological and synthetic light-harvesting complexes are also exciting opportunities.

Reproducibility and data deposition

Dual-comb spectroscopy offers the potential for exceptionally high reproducibility, precision and accuracy, owing to the intrinsic frequency calibration provided by frequency combs. In practice, systematic instrumental and sample-related effects are often the dominant limitations. Improving reproducibility therefore requires both careful experimental control and transparent reporting.

To facilitate meaningful comparison between experiments and laboratories, we suggest consistently reporting several key parameters. These include the signal-to-noise ratio, for which the definition used must be explicitly stated. We suggest defining it as the inverse standard deviation of the normalized absorption baseline constructed from the comb line maxima in spectra with resolved comb lines and reporting both the average value over a specified bandwidth (for example, 20 dB) and the peak value. Measurement time should also be specified, including both the total experimental duration and the effective data-acquisition time contributing to the interferogram, particularly when apodization or spectral interleaving is used. In addition, coherence time of the combs and the maximum averaging time over which the signal-to-noise ratio improves with the square root of time should be reported. The criteria used to define spectral resolution must be stated, along with information on comb stability under the measurement conditions. Finally, any phase-correction or resampling procedures applied to interferograms or spectra should be described, including their computational cost and potential impact on systematic errors. Adopting these minimum reporting standards would enhance transparency, reproducibility and the long-term technical maturity of dual-comb spectroscopy, particularly for applications requiring rigorous quantitative accuracy.

There are currently no community standards for the deposition of dual-comb spectroscopy data, which hinders the ability to validate and reuse data. To address this, we suggest that researchers share their raw and processed spectral data as well as their codes for data processing as additional material along with their publications or in dedicated repositories such as GitHub or Zenodo. This initiative would not only facilitate data sharing and comparison but also promote transparency and reproducibility. By adopting these practices, the community can ensure that dual-comb spectroscopy continues to provide reliable and reproducible data, supporting advancements in a wide range of scientific and industrial applications.

Limitations and optimizations

A fundamental limitation of dual-comb spectroscopy is the trade-off between sensitivity, span and measurement time. The higher the number of comb lines, the longer the measurement time to reach a given signal-to-noise ratio. This is a consequence of multiplex measurements, in contrast with multichannel parallel recording. Therefore, the span needs to be adapted to the scientific goals. For the highest signal-to-noise ratio, a comb of a few hundreds of lines may be better suited, whereas for a broad survey of many different transitions, long

measurement times or reduced signal to noise ratio are to be expected. Nevertheless, dual-comb spectroscopy remains strongly complementary to experiments of tunable laser spectroscopy. If maximum sensitivity for a single spectral line is sought, tunable laser spectroscopy may be the most suitable approach. This technique concentrates all the optical power into one frequency at a time and allows straightforward implementation of several solutions to further enhance sensitivity. Conversely, as a multiplex technique, dual-comb spectroscopy measures all spectral elements simultaneously, which grants a great consistency and precision of the spectra and, in most occurrences, reduces experimental time compared with tuning a continuous-wave laser over a broad span. To mitigate the loss in sensitivity and increased measurement time that arise when optical power is distributed over spectral regions devoid of transitions of interest, these regions can be filtered out. In dual-comb spectroscopy, experimental demonstrations have shown that combs can be spectrally tailored to emit only within the narrow spectral regions of interest⁸⁹. Spectrally dispersing the interferogram onto a detector array has been proposed¹⁶⁶ and implemented¹⁶⁷. In the dispersive configuration, using two combs with incommensurate repetition frequencies may reduce the measurement time^{26,168,169}. This approach was also initially proposed as a means of easing the instrumentation requirements for two identical laser systems^{170,171}.

Dual-comb spectroscopy works best at fixed nominal resolution – that of the comb repetition frequency. When the repetition frequency of the combs and the width of the spectral features mismatch, dual-comb spectroscopy is no longer as powerful. This is a limitation for most comb generators whose line spacing is not adjustable over several orders of magnitude, requiring the interferometer to be designed for a given range of resolutions. If a much higher resolution than the line spacing is required, a set of spectra taken with different comb line positions need to be interleaved. The multiplex character of the technique is lost, as not all spectral elements are measured simultaneously. For some comb generators, it can be a time-consuming process to repetitively selectively tune the comb lines, as it can be with a tunable continuous-wave laser. Non-resonant electro-optic combs offer adjustable repetition frequencies, but the number of comb lines is generally limited to fewer than 1,000 (refs. 38,74). Phase-modulation of combs of fixed repetition frequency has been demonstrated once to lower the repetition frequency at will but is only practicable in spectral regions where fast phase modulators are available¹⁷². Pulse picking at an adjustable picking rate also lowers the repetition frequency at will and is fully compatible with current technology for high-power amplifiers, making it attractive for exploring new spectral territories²⁶.

By contrast, if the width of the spectral features is much broader than the comb line spacing, the free induction decay is measurable only over a limited fraction of the interferogram, resulting in time spent scanning the optical delay without acquiring any meaningful signal. For example, when interrogating broad condensed-matter transitions with oscillators of 100 MHz repetition frequency, the duty cycle of measurement <1%⁴⁶. Progress has been achieved in developing and using mode-locked lasers of a repetition frequency on the order of 1 GHz, leading to refresh rates one hundred times faster than with combs of 100-MHz repetition frequency^{140,169,173}. Often, electro-optic combs do not have a sufficient span to interrogate broad spectral features^{38,74}. Alternatives can involve rapidly switching the repetition frequency of one of the combs¹⁷ or stepping the phase delay between two combs of identical repetition frequencies¹⁷⁴, but these approaches come with an additional complexity. Combs of native large line spacing such as microcombs still require optimization as discussed below.

A key technical challenge affecting any dual-comb experiment that involves nonlinear effects – whether for generating light through nonlinear frequency conversion or inducing nonlinear phenomena in a sample – stems from the low pulse energy of near-infrared frequency-comb generators operating at repetition rates above 50 MHz. A pulse energy on the order of 1 nJ is low compared with what can be reached at the output of amplified laser system of low-repetition frequency, which have been used in nonlinear optics and nonlinear spectroscopy. Therefore, the extension to frequency-comb and dual-comb techniques of the schemes that have been widespread with femtosecond chains is not straightforward. For spectroscopy in condensed matter, frequency combs of high repetition frequency would work best but they feature even lower pulse energy, whereas their average power contributes to photo-damaging the samples. For spectroscopy in the gas phase, energies on the scale of nanojoules are small for harnessing nonlinear phenomena in molecules. For nonlinear optics, nanophotonics offers interesting potential for nonlinear frequency conversion at low pulse energy. For nonlinear spectroscopy, frequency combs of incommensurate repetition frequencies^{168,175} and photon-counting techniques^{119,132} provide directions worth investigation.

The technique remains expensive to implement, requires a broad set of competences in multidisciplinary fields such as frequency metrology, spectroscopy, data processing and as any technique for high resolution or precise measurements, is full of technical and conceptual pitfalls and subtleties. Continuing to identify the optimal conditions under which dual-comb interferometry demonstrates performance advantages is essential for advancing the technology.

Improving laser sources for comb generation, particularly in the terahertz and 8–20 μm regions, is a key area of optimization. Enhanced approaches in these spectral regions can lead to better performance and wider applicability of dual-comb spectroscopy. Optimizing measurement schemes is crucial for reducing the potential for photo-damage to molecular samples in condensed phases. For instance, the optical triggering method could be a valuable approach to minimizing such damage while maintaining the integrity of the samples during measurements. The complexity of the instruments used in dual-comb spectroscopy is a challenge. Streamlining these systems can lead to more accessible and user-friendly implementations, facilitating broader adoption of the technology. Dealing with the large amount of data generated in certain implementations, such as hyperspectral imaging and time-resolved spectroscopy, requires dedicated techniques for computation and analysis. Effective data handling strategies are essential for extracting meaningful insights from complex datasets. Systematic effects in dual-comb spectroscopy have been mostly understudied. There is a need for more quantitative research to better understand and mitigate these effects, thereby improving the accuracy and reliability of the technique.

Outlook

Over the past two decades, dual-comb spectroscopy has matured and evolved. Although its potential for enhanced broadband spectroscopy has largely been realized, addressing challenges in instrumentation and applications is crucial to surpass the current state of the art and bring broadband spectroscopy into a new era across diverse scientific fields.

Instrumentation

The development of dual-comb spectroscopy instrumentation is expanding into new spectral territories, including the UV range. Although progress has been achieved in the mid-infrared and terahertz

Glossary

Absolute frequency

The measurement of an optical frequency referenced to a primary standard, enabling direct traceability to the second in the International System of Units (SI).

Accuracy

The closeness of a measured spectral parameter, such as a line position, to its true or reference value.

Aliasing

The distortion that occurs when spectral components above half the sampling rate are folded into lower frequencies owing to insufficient sampling.

Amplitude spectrum

The representation of a signal showing the amplitudes of its spectral components as a function of frequency.

Apodization

Application of a weighting (window) function to a measured interferogram, arising either physically from a finite optical delay excursion or numerically during data processing — to suppress sidelobes and reduce spectral ringing caused by truncation.

Beat note

A radio-frequency signal produced by the interference of two optical modes with slightly different frequencies.

Coherent

In which the phase of a wave or oscillation remains well defined and predictable over time or space.

Complex spectrum

A spectral representation that retains both amplitude and phase information, typically expressed as a complex-valued function of frequency.

Doppler width

The spectral broadening of a transition caused by the thermal motion of atoms or molecules, resulting in a distribution of Doppler shifts. Doppler broadening is often the dominant line-broadening mechanism in the gas phase at low pressure and room temperature.

Down-conversion

The process by which high-frequency optical signals are translated to lower radio frequencies through heterodyne detection.

Instrumental line shape

The spectral transfer function of the spectrometer, describing how an infinitesimally narrow spectral feature is mapped onto the measured spectrum by the spectrometer's resolution, sampling and coherence properties.

Interferogram

The time-domain signal produced by the interference of two optical fields whose relative phase is varied via an optical delay and which is Fourier transformed to obtain the spectrum.

Interleaving

A technique in which multiple spectra acquired with slightly shifted sampling grids are combined to increase spectral resolution and sampling density.

Mutual coherence

The degree to which the relative phase between two frequency combs evolves in a known (ideally linear) and reproducible manner in time, so that a well-defined interferogram can be measured and Fourier transformed into a spectrum.

Mutual coherence time

The time interval over which the relative phase between two frequency combs evolves in a known and reproducible manner, so that a well-defined interferogram can be recorded and Fourier transformed into a spectrum.

Nyquist condition

The requirement that a signal be sampled at a rate at least twice its highest frequency component to avoid aliasing.

Phase noise

Fluctuations in the phase of a laser field that broaden its spectral lines and limit coherence.

Phase spectrum

The representation of a signal showing the phase of its spectral components as a function of frequency.

Precision

The repeatability of a measured spectral parameter, determined by the statistical dispersion of repeated measurements.

Up-conversion

The transfer of a signal from lower to higher frequencies, typically achieved through nonlinear mixing or modulation.

Zero-filling

The addition of zeros to the end of a time-domain signal before Fourier transformation to interpolate the spectrum.

regions, the exploration of the short-wavelength range has only started^{51,52,119–123,176,177}. Specific challenges include developing UV frequency combs of low phase noise and sufficient power and achieving mutual coherence at short wavelengths. Pushing comb sources into the UV range (180–300 nm) will open doors to direct rovibronic transitions in many molecules, enabling high-sensitivity detection of air pollutants. At shorter wavelengths, this expansion promises to unlock new possibilities in precision measurements in small molecules but may require mastering dual-comb generation through high harmonic generation in rare gases^{123,178,179}. Concurrently, the use of frequency combs with both high and low-repetition frequencies (compared with the common hundreds of megahertz) is essential for addressing a wide range of new applications. This includes achieving very high resolution for precision measurements²⁶ and very low resolution for condensed matter spectroscopy⁴⁶.

Dual-comb spectroscopy has the potential of pushing the boundaries of precision spectroscopy over broad spectral bandwidth and of enabling new forms of quantum-enhanced spectroscopy. Advancement

in instrumentation for the detection and analysis of weak signals will enable extremely accurate and sensitive measurements, such as in fundamental physics and advanced material research.

Further progress in ruggedized, battery-powered dual-comb instruments for open-air monitoring of pollutants and greenhouse gases, for example, will be transformative for climate research, industrial compliance and urban air-quality networks. Bringing dual-comb functionality onto a monolithic photonic platform will not only shrink size, weight, power and cost but also allow mass-manufacturable, rugged spectrometers for field use — from handheld environmental sensors to embedded industrial monitors⁹¹. The practical consequence of the dual-comb interferometer performing direct time–frequency domain measurements without being constrained by geometry is that spectrometers can be made arbitrarily compact^{12,25}. Determining how compact these systems can become and whether a complete gas-phase dual-comb spectroscopy laboratory can be implemented on a photonic chip are among the most exciting challenges in instrumental development.

Emerging applications

In laboratory spectroscopy, dual-comb spectroscopy holds largely unexplored potential – particularly for achieving Doppler-free spectroscopy over broad spectral bandwidths^{26,52}. In this area, it introduces capabilities that are entirely new to broadband spectroscopy: exceptionally high resolution in a single acquisition, surpassing the fundamental limits imposed by optical aberrations, and direct frequency-scale calibration. A key challenge and opportunity will be to determine how far its accuracy can be advanced and whether dual-comb spectroscopy can evolve into a broadband frequency metrology tool – one capable of benchmarking ab initio quantum chemistry calculations and contributing to the determination of fundamental constants.

For environmental sensing, expanding the use of dual-comb spectroscopy for high-resolution, open-path measurements of reactive species (such as NH₃, NO_x and volatile organic compounds) in the complex and dynamic environments of urban atmospheres, agricultural sites and combustion systems, is an exciting direction. Although dual-comb spectroscopy has been used in long-path greenhouse gas sensing, pushing towards multi-kilometre, all-weather, continuous monitoring of trace gases with parts per billion-level sensitivity remains a major scientific goal, requiring advances in mid-infrared comb power, detector sensitivity and atmospheric noise mitigation. Another emerging frontier is hyperspectral dual-comb spectroscopy imaging of gas plumes and surfaces, which could enable spatial mapping of leaks or emissions in real time. This requires rapid, sensitive and pixel-resolved detection schemes.

More generally, extending dual-comb methods into two-dimensional and three-dimensional imaging modalities – combining high spectral resolution with spatial mapping – offers enhanced capabilities for capturing detailed spectral and spatial information and could impact other applications ranging from material diagnostics to medical diagnostics, such as in label-free tissue mapping. Dual-comb spectroscopy is paving the way for advancements in hyperspectral imaging and holography.

The rapid development of dual-comb spectroscopy is expected to accelerate its application in condensed-phase physics, chemistry and biology. For example, dual-comb and multi-comb spectroscopy offer prospects for pump-probe, two-colour pump-probe and multidimensional spectroscopy by enabling direct, scan-free access to ultrafast dynamics with high spectral resolution and phase stability. In pump-probe schemes, dual-comb approaches can allow rapid acquisition of time-resolved spectra with femtosecond to nanosecond resolution, which is especially promising for studying nonequilibrium dynamics in complex systems such as molecular aggregates, solids or reactive intermediates. In multidimensional spectroscopy, multi-comb configurations could permit simultaneous control over multiple time delays, enabling full electric-field-resolved two-dimensional or three-dimensional spectroscopic measurements without moving parts. However, key challenges remain: long coherence time usually involves single systems or dilute samples requires achieving sufficient optical power and pulse shaping flexibility, particularly in the mid-infrared and UV where many fundamental excitations lie, is essential. Overcoming such hurdles could open new regimes of coherent spectroscopy with unprecedented speed and information content.

Dual-comb spectrometers have matured into versatile, multidisciplinary tools that can be effectively integrated with complementary technologies. The future of this emerging field will be shaped by the ingenuity and innovation of the scientists driving its development.

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Competing interests

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