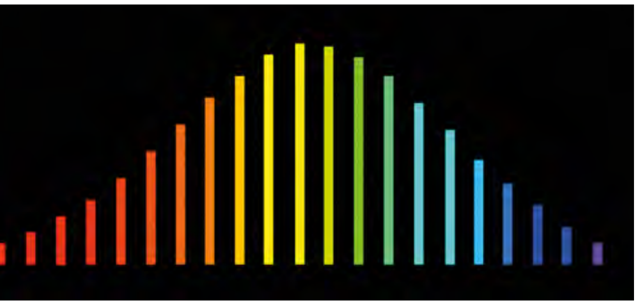
INTERFEROMETRY
WITH OPTICAL FREQUENCY COMBS

Nathalie PICQUÉ[1,*], Theodor W. HÄNSCH[1,2]
[1]Max-Planck Institute of Quantum Optics, Garching, Germany
[2]Ludwig-Maximilian University of Munich, Faculty of Physics, Munich, Germany
***nathalie.picque@mpq.mpg.de**



https://doi.org/10.1051/photon/202111338

A frequency comb, a spectrum of equidistant phase-coherent laser lines, can be harnessed for new approaches to interferometry. The dual-comb interferometer exploits the time-domain interference between two combs of slightly different line spacing. The instrument, which performs direct frequency measurements over a broad spectral bandwidth, opens up new perspectives in applications such as spectroscopy, distance metrology or holography.

A frequency comb [1] is a spectrum of narrow evenly spaced laser lines, whose absolute frequency can be known within the accuracy of an atomic clock (Insert 1). Initially invented for precision frequency metrology in the simple hydrogen atom, frequency combs have become key to a variety of applications, from the generation of attosecond pulses to the calibration of astronomical spectrographs. They are enabling ground-breaking approaches to interferometry with applications as diverse as spectroscopy [2], distance metrology [3], or holography [4]. This short article recounts the principle and applications of frequency comb interferometry.

FREQUENCY COMBS AND INTERFEROMETRY

Early on, frequency combs have been coupled to known interferometers. For example, if a frequency comb is used as a light source before a scanning Michelson interferometer, Fourier transform spectroscopy sees its measurement speed, sensitivity precision and accuracy dramatically improved. Distance measurements using a spectrally-dispersed static Michelson interferometer -or a scanning Michelson interferometer- benefits from the many comb lines for improved precision and large ambiguity range. A comb of narrow line spacing can be frequency filtered in a Fabry interferometer, of matching but larger free spectral range, to generate a comb of large repetition frequency, suited to the calibration of astronomical spectrographs. The Fabry-Pérot resonator can also be used as an enhancement cavity, for extreme-ultraviolet high-harmonic generation or for absorption spectroscopy with long absorption paths.

DUAL-COMB INTERFEROMETERS

More interestingly, frequency combs have enabled a new class of interferometers, called dual-comb interferometers. Two frequency comb generators emit trains of pulses at slightly different repetition frequencies, f_{rep} and $f_{rep}+\Delta f_{rep}$, respectively. The two 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.1a), the pulses from one laser walk through the pulses from the second laser, with a time separation that automatically increments from pulse pair to pulse pair by an amount $\Delta f_{rep}/ f_{rep}^2$ (e.g. 10^{-14} s). This way, optical delays from 0 to $1/f_{rep}$ are repetitively scanned without moving parts. Akin to a sampling oscilloscope, the periodic optical waveforms are stretched in time by a factor $f_{rep}/\Delta f_{rep}$ (e.g. 10^6), and they can be electronically recorded and digitally processed. In the frequency domain (Fig.1b), pairs of optical comb lines, one from each comb, produce radio-frequency beat notes on the detector, forming a frequency comb of line spacing Δf_{rep} in the radio frequency domain. Optical frequencies $nf_{\mathrm{rep}}+f_0$ are thus down-converted into radio frequencies $n\Delta f_{\mathrm{rep}}+\Delta f_0$.

The principle of dual-comb interferometry may be reminiscent of that of asynchronous optical sampling. A significant difference, though, is that mutual coherence between the two combs is required for an interferometric measurement. As a rule of thumb, many applications require a relative stability on the order of $\lambda/100$ to achieve high fringe contrast. At a wavelength of 1 µm, this means that the timing jitter between pairs of pulses must be kept smaller than 30 as. Learning how to control and minimize the relative timing and phase fluctuations between two frequency comb generators has been at the center of significant efforts in the early days of dual-comb interferometry. A number of solutions, of various complexity and performance, now enable to experimentally maintain mutual coherence and/or to compensate for the (residual) fluctuations through analog or digital processing. Experimentally, coherent averaging over more than half an hour has become feasible, illustrating the degree of control achieved for a dual-comb system.

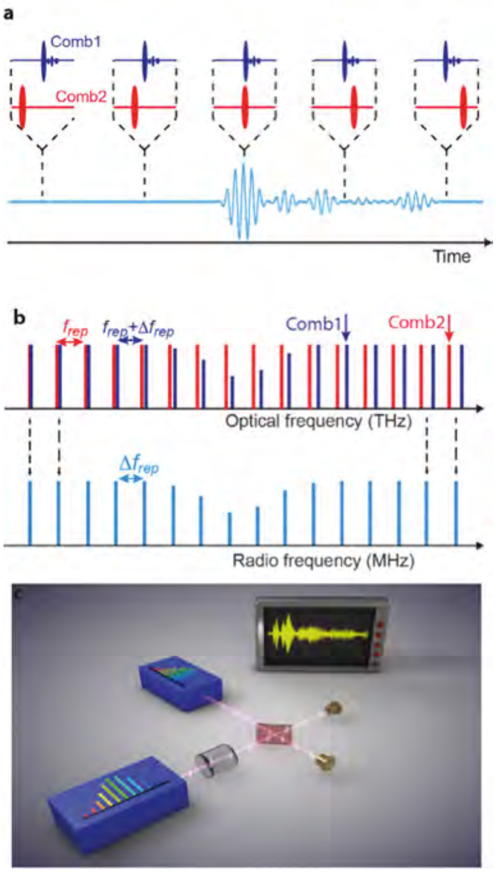
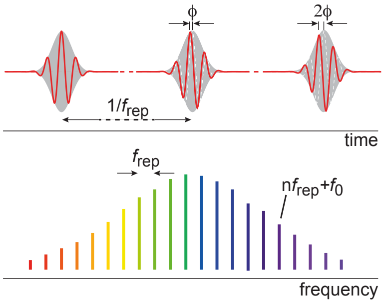


Figure 1. a. Time-domain principle of dual-comb interferometry (in the situation where an absorbing sample is in the beam path of comb 1). **b.** Frequency-domain principle **c.** Sketch of a typical dual-comb spectrometer. **c.**: Adapted from "Mid-IR Spectroscopic Sensing," Optics & Photonics News **30(6)**, 26-33 (2019). https://doi.org/10.1364/OPN.30.6.000026

FREQUENCY COMB



A frequency comb, a spectrum of equidistant phase-coherent spectral lines, is commonly generated by a mode-locked laser. Such a laser emits a periodic train of pulses at a frequency f_{rep}. The periodicity applies not only to the pulse envelopes, but to the whole electric field of the pulses, including their optical phase, apart from a reproducible slip ϕ of the phase of the electromagnetic carrier-wave relative to the pulse envelope from pulse to pulse. Such phase slips occur in a laser owing to dispersion in the cavity. As a consequence, the frequency f_n of a comb line writes: $f_n = n\, f_{rep} + f_0$ where n is an integer and $f_0 = f_{rep}\,\phi/\ 2\pi$ is the carrier-envelope offset frequency. Both f_{rep} and f_0 can be measured and controlled against an atomic clock. Therefore, frequency combs act like rulers in frequency space that can for instance be used to measure a large separation between two different optical frequencies in terms of the countable signal of the pulse repetition frequency. Frequency combs can conveniently link optical and microwave frequencies and enable absolute measurements of any frequency. Frequency combs have revolutionized the way the frequency of light is measured and are now common equipment in all frequency metrology-oriented laboratories. They have paved the way for the creation of all-optical clocks with a precision that approaches the 10^{-18} level.

Owing to the absence of moving parts, one of the first features of dual-comb interferometers that attracted interest has been the ability to perform fast measurements. Moreover, the sensitivity is further enhanced. is further enhanced by the use of coherent light sources. Remarkably, a dual-comb interferometer can simulate a mirror moving at 10 $km.s^{-1}$, the escape velocity from earth. The beat notes between pairs of comb lines are mapped in the radio-frequency domain where the 1/f noise in the detector signal can be greatly reduced, and a million-fold improvement in the acquisition speed of a spectrum has been demonstrated. Furthermore, all the spectral elements are simultaneously measured on a single photodetector, like in other interferential spectrometers. This multiplexed recording grants excellent overall consistency of the spectral measurements and applicability in any spectral regions. As research progressed, other distinguishing assets have been highlighted, offering interferometry a unique and novel host of powerful features, which are summarized below for the key applications.

DUAL-COMB SPECTROSCOPY

The main application of dual-comb interferometers has been spectroscopy over broad spectral bandwidths, in particular of molecules [2]. Dual-comb spectroscopy is a new form of Fourier transform spectroscopy, which has been over the past 50 years the overriding tool in molecular spectroscopy and analytical chemistry. If a sample is present on the beam path of one of the combs (Fig.1c), the interference signal will record the signature of the absorption and dispersion experienced by the sample. The dual-comb spectrometer mimics a scanning Michelson interferometer by sampling the free-induction of the molecules over the range of optical delays. One uses then a harmonic-analysis tool, the Fourier transformation, to get the spectrum. In a single recording, the spectrum is sampled by the discrete comb lines and therefore the spectral resolution is limited to the comb line spacing f_{rep}. As most combs can be precisely controlled, it is however possible to measure a sequence of spectra with different comb-line positions and to interleave the spectra a posteriori to reach a resolution ultimately limited by the width of the optical comb lines. Initially developed in the near-infrared spectral region, where frequency comb generators are conveniently available, dual-comb spectroscopy is now efficient in the mid-infrared spectral region at wavelengths as long as 5 µm (Fig. 2). It is emerging at longer wavelengths in the mid-infrared and THz ranges and at shorter wavelengths in the visible range. The ultraviolet domain is still mostly unexplored, due to outstanding instrumental challenges for generating low-noise frequency combs of a broad span and implementing ultra-stable interferometers at short-wavelengths.

To broadband spectroscopy, dual-comb interferometers add the remarkable features of the interrogation of the sample by laser lines of narrow width which provides a negligible contribution of the instrumental line-shape, as well as the calibration of the frequency scale within the accuracy of an atomic clock. These features are not available from any dual-comb interferometers though. They derive from the use of frequency combs of narrow and stable optical lines, referenced to a radio-frequency clock or to an ultra-stable optical frequency standard. Initially, it was not clear which light sources should be used and a variety of comb generators have been tested, from the metrology-grade fiber mode-locked lasers stabilized to accurate optical references to simple electro-optic modulators or quantum cascade lasers that are free-running. Over the years, the fiber-laser technology has considerably evolved towards ease of use, compactness, low noise and ultra-high stability. Fiber laser have become the most suited tools for realizing the potential of dual-comb interferometry. Free-running frequency combs do not enable to benefit from the above-mentioned advantages, although they might still be valuable tools in some niche applications.

Although linear absorption spectroscopy has been predominantly explored, frequency combs synthesizers based on mode-locked lasers involve intense ultrashort pulses that can generate nonlinear phenomena at the sample. Using this, various schemes of nonlinear Raman spectroscopy and imaging and of Doppler-free two-photon excitation spectroscopy [5] have open up new opportunities for nonlinear spectroscopy over broad spans. One can expect that this line of research will develop further in the near future, owing to the progress in high-power laser amplifiers at high repetition frequency. Finally, the system is not limited to two frequency combs and recent proof-of-principle demonstrations explore the intriguing potential of using three combs for multidimensional spectroscopy, such as photon echoes [6].

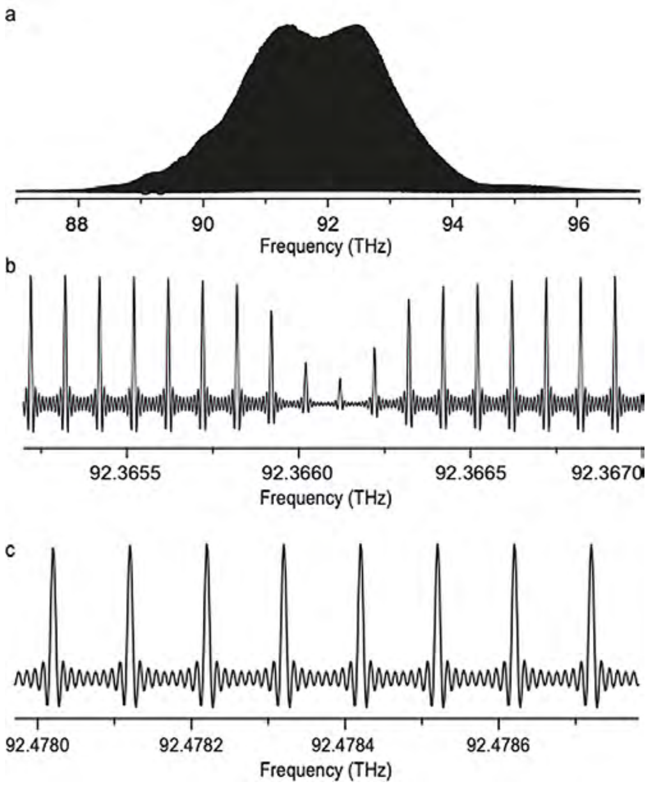


Figure 2. Mid-infrared experimental dual-comb spectrum, shown with different magnifications. **(a.-c.)** 82000 comb lines spaced by 100 MHz, centered at 92 THz (3.2 µm), were measured within 29 minutes. In **b**. an absorption transition of ethylene attenuates the comb lines. Reproduced from "Mid-infrared feed-forward dual-comb spectroscopy," Proc. Natl. Acad. Sci. USA **116**, 34549 (2019). https://doi.org/10.1073/pnas.1819082116

DUAL-COMB RANGING AND HOLOGRAPHY

Another successful application of dual-comb interferometers involves distance measurements and ranging [3]. The dual-comb scheme combines the time-of-flight and interferometric approaches to deliver absolute distance measurements over an extended ambiguity range.

One of the most exciting recent trends, though, has been to replace the single photodetector by a camera sensor. As many spectra as there are detector pixels can be simultaneously measured. Dual-comb interferometers move digital holography ● ● ●

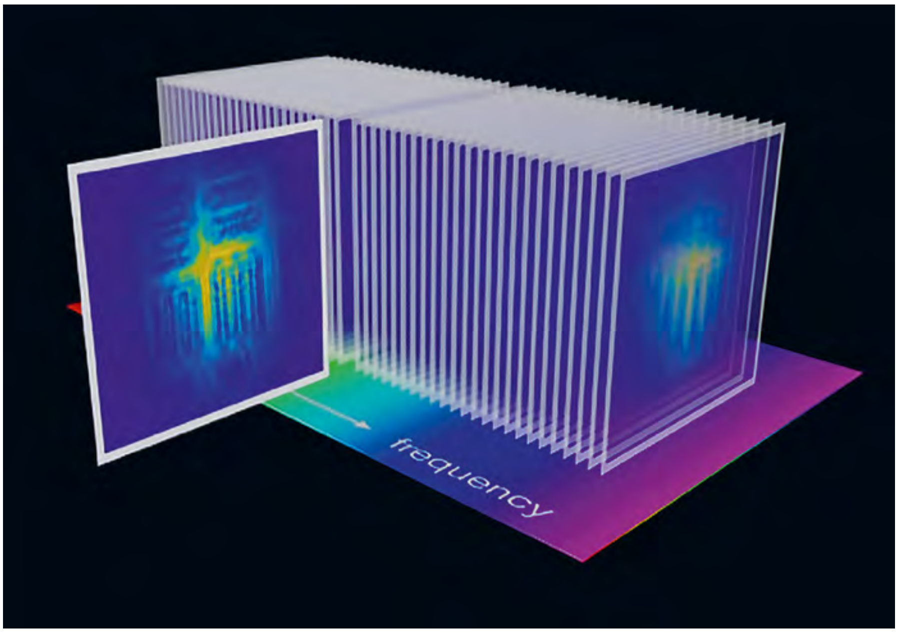


Figure 3: In dual-comb holography, as many holograms as there are comb lines are generated.

forward by recording simultaneously thousands of holograms [4], one per comb line (Fig.3), and they show an intriguing potential for reaching new frontiers in scan-free wavefront reconstruction. By digital processing, each hologram provides a 3-dimensional image of the scene, where the focusing distance can be chosen at will. Combining all these holograms renders the geometrical shape of the 3-dimensional object with very high precision and without ambiguity. At the same time, other diagnostics can be performed by the frequency combs: in the first proof of concept, molecule-selective imaging of a cloud of ammonia vapor was simultaneously demonstrated.

CONCLUSION

Dual-comb interferometry has been developed over the past 15 years and now involves a vibrant research community of more than 200 research groups. The dual-comb interferometer provides a unique combination of broad-spectral-bandwidth, long temporal coherence, absence of moving parts and multi-heterodyne read-out which offers interferometry a revolutionary host of features – frequency multiplexing, resolution, accuracy, precision, speed. Other emerging applications include analog-to-digital conversion, two-way time and frequency transfer, vibrometry, etc.

Excitingly, the technique is still far from realizing its full potential. A remarkable difference with respect to other types of interferometers is that the dual-comb interferometer performs direct time/frequency measurements. With a dispersive or interferential instrument, the resolving power is the ratio of the maximum path difference to the wavelength, hence the bulky instruments for high resolution. With a dual-comb interferometer, it 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. This apparently simple but fundamental distinction, first pointed out in [2], has not been exploited yet in experiments that would go significantly beyond the state of the art. On an applied touch, the path is open to integrated chip-scale ultra-miniaturized devices that combine high resolution and broad spectral bandwidth. Using III-V-on-silicon mode-locked lasers in the telecommunication region, an on-chip spectroscopy laboratory for gas sensing is already underway using battery-operated devices of a footprint smaller than 1 mm^2 [7]. The most exciting prospect is nevertheless that of merging frequency metrology and broadband spectroscopy: with Doppler-free dual-comb spectroscopy, the envisioned improvement in accuracy is expected similar to that achieved in the 1990s when going from optical wavelength metrology to frequency measurements. As the technique is able to measure very faint signals, down to the single-photon level, single atoms and molecules may become observable over a broad span with an unmatched precision, opening up new strategies for tests of fundamental physics. ●

REFERENCES

[1] T. Udem, R. Holzwarth, T.W. Hänsch, Nature **416**, 233 (2002)

[2] N. Picqué, T.W. Hänsch, Nat. Photon. **13**, 146 (2019)

[3] I. Coddington, W.C. Swann, L. Nenadovic *et al.* Nat. Photon. **3**, 351 (2009)

[4] E. Vicentini, Z. Wang, K. Van Gasse *et al.* Nat. Photon. **15**, 890 (2021)

[5] S.A. Meek, A. Hipke, G. Guelachvili *et al.* Opt. Lett. **43**, 162 (2018)

[6] J.W. Kim, J. Jeon, T.H. Yoon *et al.*, J. Opt. Soc. Am. B **39**, 934 (2022)

[7] K. Van Gasse, Z. Chen, E. Vicentini, *et al.* preprint at arXiv:2006.15113 (2020)