Takuro Ideguchi

Dual-Comb Spectroscopy

Using paired coherent frequency combs for broadband molecular spectroscopy can provide dramatic gains in spectral resolution, sensitivity and data acquisition speed—and may help take the power of frequency comb spectroscopy out of the lab and into the field.
Dual-Comb Spectroscopy

Using paired coherent frequency combs for broadband molecular spectroscopy can provide dramatic gains in spectral resolution, sensitivity and data acquisition speed—and may help take the power of frequency comb spectroscopy out of the lab and into the field.
Pairing objects can bring new functionality. A pair of chopsticks easily feeds us, while a single one would be useless. A pair of eyes lets us visualize the world in 3-D, rather than the flat, 2-D view we would have with only one.

In a similar way, research in precision spectroscopy over the past decade is demonstrating that pairing optical frequency combs can open up new functionality and applications. Honored with the 2005 Nobel Prize in physics, broadband frequency comb spectroscopy—in which the signal from an optical frequency comb is read by a conventional spectrometer—has already enabled measurements of atomic and molecular structures at high resolution. But in the technique called dual-comb spectroscopy, that conventional spectrometer—and that instrument’s limitations on speed and resolution—are removed. Instead, a second frequency comb takes on the work previously done by the spectrometer.

The result can be dramatic gains in data acquisition speed, spectral resolution and sensitivity, which are starting to show up in both lab demonstrations and field applications. This feature offers an overview of this emerging technique, some recent research advances, and future prospects and applications.

**Optical frequency combs**

An optical frequency comb is a spectrum consisting of hundreds of thousands or millions of equally spaced, sharp lines—analogous to having a great many continuous-wave (CW) lasers simultaneously emitting at different, equally spaced frequencies. Optical combs can be generated in many ways; the most common method uses a phase-stabilized, mode-locked ultrashort-pulse laser. In the time domain, the laser produces a pulse train at a specific repetition rate, and with a specific, increasing additional carrier-envelope phase with each successive pulse. When the repetition rate and carrier-envelope phase of the pulse train are both stabilized against radio- or optical-frequency references, a Fourier transformation of the laser’s periodic pulse train shows a sharp, comb-like spectrum in the frequency domain.

If the frequency comb is well stabilized and referenced to an absolute frequency standard, such as an atomic clock, the comb spectrum becomes an extremely precise ruler for measuring optical frequencies. That ruler has found applications in a wide variety of scientific problems: high-resolution frequency measurements of atomic, ionic or molecular transitions to answer fundamental questions in physics; the detection of tiny amounts of Doppler shift in hunting extrasolar planets; and other applications in attosecond physics, ultrapure microwave generation, time–frequency transfer over long distances, manipulation of atomic qubits, and atomic-clock intercomparison, to name a few.

One of the most active research areas for frequency combs is broadband molecular spectroscopy. The comb’s millions of equally spaced, sharp lines offer the opportunity to measure complex broadband molecular signatures with high spectral resolution and sensitivity. Exploiting those advantages, however, requires a spectrometer of sufficiently high resolution to resolve each individual comb line. One approach uses a spectrometer based on a virtually imaged phased array (VIPA) disperser in combination
Because the dual-comb setup does not depend on the mechanical motion of a mirror, its scanning rate is several orders of magnitude faster than that of the Michelson-type interferometer.

with a diffraction grating; another common scheme uses a workhorse of analytical chemistry, the Michelson-type Fourier transform spectrometer, and replaces the conventional broadband, usually incoherent light source with a frequency comb.

In this approach to frequency comb spectroscopy, the frequency comb pulse train is split into interferometer arms, one of which includes a mechanically scanned mirror, and the two pulse trains are sent through the sample to be analyzed. As the mirror is scanned, a series of interferograms is recorded with a single photo-receiver and a digitizer; Fourier transformation of the interferograms generates the spectrum, with a resolution determined by the maximum optical-path-length difference of the interferometer.

The dual-comb advantage
A key drawback of doing frequency comb spectroscopy with the Michelson-type setup outlined above is speed: the scan rate of the setup, which is limited by the velocity of the scanning mirror, is commonly only on the order of Hz. Dual-comb spectroscopy seeks to get around that disadvantage by using a second frequency comb, rather than a moving mirror, to supply the delay time. The result can be a significant enhancement of the spectrometer’s performance.

In the dual-comb setup, the pulse train forming a second comb, with a slightly different pulse repetition rate from the first, is spatially combined with the train from the first comb. The combined pulse train is passed through the sample to be analyzed, and detected by a photo-receiver. The result, in the time domain, is a repeated series of cross-correlation-like interferometric signals between the pulses, with a steadily faster than that of the Michelson-type interferometer.
To get dual-comb spectroscopy out of the lab and into practical use will require a way to simplify the comb stabilization, yet retain the quality and speed of the technique.

increasing time difference based on the difference in repetition rate between the two combs.

The dual-comb interferograms thus have characteristics similar to those of a conventional Michelson-type Fourier transform spectrometer—but, because the dual-comb setup does not depend on the mechanical motion of a mirror, its scanning rate is several orders of magnitude faster than that of the Michelson-type interferometer. Moreover, the dual-comb interferometer can provide high spectral resolution, because the pulse repetition rate determines the maximum optical delay, which in turn determines the instrumental spectral resolution. Combs with a repetition rate of around 100 MHz, for example, can generate a maximum delay of 10 ns, which corresponds to a 1.5-m linear optical delay line at atmospheric conditions. Keeping a mechanical delay line stable for that distance, within interferometric precision, is not a trivial task.

Another advantage of dual-comb spectroscopy emerges in the frequency domain. There, the mixing of the two optical combs, with slightly different repetition rates, results in a third, down-converted radio-frequency (RF) comb, with spacing between teeth equivalent to the repetition rate difference between the two optical combs. The sample’s response is thus encoded on this down-converted RF comb, and the beat measurement between the two optical combs generates a multi-heterodyne signal that can be recovered from the RF comb. In sum, the down-converted comb inherits the coherence property of the optical frequency combs, enabling broadband spectroscopy with a high resolution and accuracy—with the speed and digital-signal-processing advantages of RF heterodyne detection.

Implementing dual-comb spectroscopy
A key component of any dual-comb system is, of course, the comb sources. Most common have been femtosecond mode-locked lasers, especially fiber lasers. The repetition rate of these lasers typically lies in the range of...
tens to hundreds of MHz, so dual-comb systems based on them are ideal for gas-phase molecular spectroscopy with Doppler-limited resolution.

Other comb sources, however, have also recently shown intriguing potential for dual-comb spectroscopy. One such source involves cascaded four-wave mixing (FWM) in microcavities, to generate, through the nonlinear Kerr effect, a so-called microcomb or Kerr comb. These comb sources can be implemented on CMOS-compatible semiconductor chips, which suggests the ability for cost-effective mass production and widespread use in compact dual-comb spectrometers. Another emerging comb source, the electro-optic modulator (EOM) comb, operates by combining intensity modulations of a single CW laser with two EOMs, inserted in branched optical paths, to generate mutually coherent dual combs. The common-mode noise cancellation guarantees a comb-resolved resolution without the need for active stabilization.

The spatial coherence of these laser combs means that one can enhance the actual signal from interaction with the sample through various techniques. One of the most straightforward is a multi-pass cell, which elongates the light-matter interaction length to boost signal quality. Although the comb itself includes a massive number of individual spectral teeth, these can simultaneously resonate in such a cavity as long as the modes of the cavity and the comb match each other within a width set by the cavity’s finesse. A dispersion-managed cavity can also enhance the effective interaction length by orders of magnitude while maintaining the frequency comb’s broad bandwidth.

The coherent nature of frequency comb lasers also enables open-path measurement through the atmosphere for remote sensing. Dual-comb spectroscopy could even prove valuable for microscopy—installing a laser scanning microscope in the dual-comb system enables a broadband spectroscopic microscope, and near-field and nonlinear dual-comb microscopy have already been demonstrated.

**Making dual-comb techniques practical**

Although dual-comb spectroscopy is simple in principle, and although new comb sources and experimental setups are emerging, practical implementation of the technique remains demanding, because of the requirement of two mutually coherent frequency combs. Many dual-comb experiments thus far have relied on fully stabilized combs with phase-locking systems with respect to both the repetition and carrier-envelope offset frequencies—setups that are difficult to replicate outside of the research lab. Yet attempting to do dual-comb spectroscopy with two free-running lasers (i.e., lasers without phase stabilizations) would lead to significant temporal distortions on the interferograms and, in turn, chromatic distortions on the Fourier-transformed spectrum.

To get dual-comb spectroscopy out of the lab and into practical use, therefore, will require a way to simplify the comb stabilization, yet retain the quality and speed of the technique. One way to simplify dual-comb spectroscopy is **real-time phase error correction**—that is, measuring the phase fluctuations between the combs at the same time the interferograms are measured, and correcting the phase error computationally.
As dual-comb technology has rapidly matured, it has started to see real-world applications beyond laboratory proof-of-concept experiments.

Real-time phase error correction is routine for Michelson-based Fourier transform spectrometry, and is achieved by installing a He-Ne laser collinearly with the infrared beam used for the spectroscopy. It has also shown effectiveness in dual-comb spectroscopy setups, though here the case is more complicated because there are two degrees of freedom to be compensated. From a computational perspective, field-programmable gate array technology can make the correction process in real-time.

An analog counterpart of such digital computational phase error correction, called adaptive sampling, has also been demonstrated. In this case, the phase error correction can be done with all passive analog electronic circuits, eliminating the need for the post-processing computational step. Using these techniques would, in principle, remove the requirement of strict phase-locking feedback systems from the dual-comb setup, requiring only looser stabilizations for long-term measurement.

An even simpler potential implementation of dual-comb spectroscopy is single-laser dual-comb spectroscopy—that is, using a single free-running laser to generate both combs. A laser cavity may emit two pulse trains at slightly different repetition rates in some laser configurations. Because such pulses share a single cavity, the pulse trains have passive mutual coherence due to the common-mode noise cancellation.

Such dual-comb lasers have been demonstrated in various types of laser cavities, including semiconductor disc lasers, monolithic cavity lasers, fiber lasers and Ti:sapphire-based solid-state lasers. The single-cavity-based dual-comb laser could prove a very satisfactory approach, as it requires neither a second laser source nor a phase-locking system.

**Dual-comb techniques in the real world**

As dual-comb technology has rapidly matured, it has started to see real-world applications beyond laboratory proof-of-concept experiments.

**Greenhouse-gas sensing.** One example, from 2014, involved an application to the study of climate change and greenhouse gas emissions. Here, a research team used a dual-comb setup involving two mutually coherent erbium-doped fiber laser comb sources, with spectra centered near the near-infrared 1.6-μm water vapor window, to perform atmospheric absorption spectroscopy via remote sensing across 2 km of free space.

Not only did the method allow for tracking of variations on the order of 1 part per million for CO₂ and 3 parts per billion for methane (both potent greenhouse gases), but the technique’s fast acquisition speed, according to the research team, rendered it immune from the common problem of intensity noise from atmospheric turbulence. Other work has demonstrated combustion gas sensing in a power-plant setting; here, a robust dual-comb spectrometer successfully measured temperature, H₂O and CO₂ concentration variations in the exhaust of a stationary natural-gas turbine.
**Molecular fingerprinting** and trace gases. The real-world experiments noted above have involved near-infrared frequency combs. Extending the advantages of dual-comb spectroscopy to a broader range of molecular species will require comb sources in the mid-infrared region—the so-called molecular-fingerprint region, in which fundamental molecular vibrations provide valuable signatures for a wide range of environmentally, chemically, or biomedically important trace gases and other molecules.

In contrast to the ready availability of near-infrared sources, however, mid-infrared combs are still in the development stage. Nonetheless, nonlinear frequency conversion techniques such as optical parametric oscillation or difference frequency generation have made it possible to convert conventional near-infrared combs into mid-infrared combs. Intensive work on stabilizing such combs has led to a proof-of-concept demonstration of dual-comb spectroscopy in the mid-infrared. Other work has investigated direct mid-infrared comb generation from laser oscillators (such as Cr-based femtosecond lasers, crystalline or silicon microresonators and quantum cascade lasers), without the need for frequency conversion. Such direct mid-infrared comb sources, if realized, could find use in simple, robust dual-comb spectrometers for molecular fingerprinting.

**Coherent Raman dual-comb spectroscopy and biomedical diagnostics.** Another way to access the fingerprint region is through Raman scattering, which can use conventional near-infrared frequency combs rather than demanding mid-infrared comb sources. Unfortunately, however, linear, spontaneous Raman scattering is a very weak phenomenon and thus cannot harness the high-speed capabilities of a dual-comb scheme.

We have found one route around that disadvantage: using coherent Raman scattering—a nonlinear Raman-scattering process that enhances the Raman signal by orders of magnitude relative to the weaker linear spontaneous process. Because femtosecond-laser-based frequency combs consist of ultrashort pulse trains, the pulses with high peak intensity can efficiently induce nonlinear optical effects. Here, the ultrashort pulse train of the comb periodically excites molecular vibrations via an impulsive stimulated Raman scattering process. The pulse train of the second comb, with a linearly increasing time delay, probes the excited vibrations.

Using this approach, we have incorporated coherent Raman scattering into the dual-comb modality, and were able to demonstrate ultra-rapid coherent Raman spectroscopy, while maintaining the dual-comb spectroscopy advantages of high resolution, sensitivity and broad bandwidth. As the system continues to be refined, dual-comb spectroscopy encompassing coherent Raman scattering could emerge as a powerful label-free, diagnostic molecular fingerprinting technique for biomedicine and materials science. Applications could include rapid, broadband spectroscopic bioimaging for live cell or tissue analysis.

**Beyond spectroscopy.** Finally, dual-comb systems offer several intriguing possibilities beyond spectroscopy. One recent example involves use of a dual-comb system with coherent LIDAR for rapid absolute distance measurement. Here, the ability to seamlessly switch from the coarse measurement allowed by time-of-flight LIDAR to the fine-scale measurement of dual-comb interferometry enables nanometer-scale resolution coupled with kilometer-scale unambiguous range. Dual-comb systems have also found uses in frequency calibration of continuous-wave lasers and in optical two-way time and frequency transfer over space. All of which shows, once again, that putting two frequency combs together can offer much more than just the sum of the parts.

---

Takuro Ideguchi (ideguchi@gono.phys.s.u-tokyo.ac.jp) is with the School of Science, University of Tokyo, Japan.

---

**References and Resources**