



Dual-Comb and Fourier Transform for Fast and High Spectral Spectroscopy

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ABSTRACT

Complex chemical and biological reactions require fast (sub-second) spectroscopy with high spectral resolution. Achieving both of these standards with commercial instruments has proven to be a challenge. Researchers from Switzerland have developed a Fourier Transform (FT) spectrometer designed around a custom, continuous rotation scanning mirror, enabling millisecond acquisition times while maintaining high spectral resolution in the sub-GHz range. The demonstrated system incorporates the advantages of both FT spectroscopy as well as dual-comb spectroscopy. This design, not only uses a single frequency comb for dual-comb spectroscopy, but can prove useful in a variety of spectroscopy applications where high speed, large optical bandwidth, and high spectral resolution are desired.

SPECTRAL AND TEMPORAL RESOLUTION

In the world of spectroscopy, real-time monitoring and analysis are crucial for understanding underlying complex dynamics. Whether in chemical reactions (**Figure 1**), protein dynamics, micro-machining, or phase transitions in novel material systems,¹ spectrometers play a major role, one requiring high temporal and high spectral resolution.

Often times, a general purpose instrument can only have fast acquisition speeds or high spectral resolution. This results in a trade-off between temporal and spectral resolution in spectrometer instrumentation. This is one of the major challenges alongside the miniaturization of spectrometers for cost-effective lab-on-a-chip systems.¹ Two methods have proven effective in spectroscopic characterization: Fourier-Transform (FT) spectroscopy and dual-comb spectroscopy.

FT spectroscopy offers a relatively high spectral resolution when transforming the frequency signals into the absorption spectrum useful for analysis. Traditional FT spectrometers utilize a mirror that is linearly displaced to obtain the information, hence the decreased temporal resolution. Dual-comb spectroscopy, especially in the mid-infrared (IR), enables extremely fast acquisition times while covering a relevant spectral range for molecular spectroscopy of 4 - 12 μm . As Mid-IR quantum cascade lasers (QCLs) continue to increase in accessibility and adaptation for spectroscopy, high spectral and temporal resolution can be achieved with the correct instrumentation and techniques.

There are a few criteria that are desirable for real-time monitoring and analysis: large optical bandwidth, high spectral resolution, preserved polarization, fast acquisition speeds, small footprint, decoupled temporal and spectral resolutions, and support of extended beam diameter size.¹



Figure 1. Chemical reactions - one of the leading applications for real-time, rapid monitoring and analysis.

PROBLEMS AND GOALS

Although FT and dual-comb spectroscopy do bring high spectral resolution and fast acquisition times, respectively, both methods have disadvantages. A traditional FT spectrometer's scan rate is too slow for most applications due to the linear movement of a mirror for information. Other methods of FT have sought to improve on the temporal resolution but usually require and depend on dispersive elements. While these changes improve temporal resolution, the spectral bandwidth is sacrificed.

When combining FT and dual-comb spectroscopy to achieve both high spectral and temporal resolution, maintaining coherence for both frequency combs is a challenge. Additional noise can be generated from the lack of mutual coherence of two free running lasers.¹ Thus it would be beneficial to achieve dual-comb spectroscopy with only a single frequency comb source without creating sacrifices of the criteria listed.

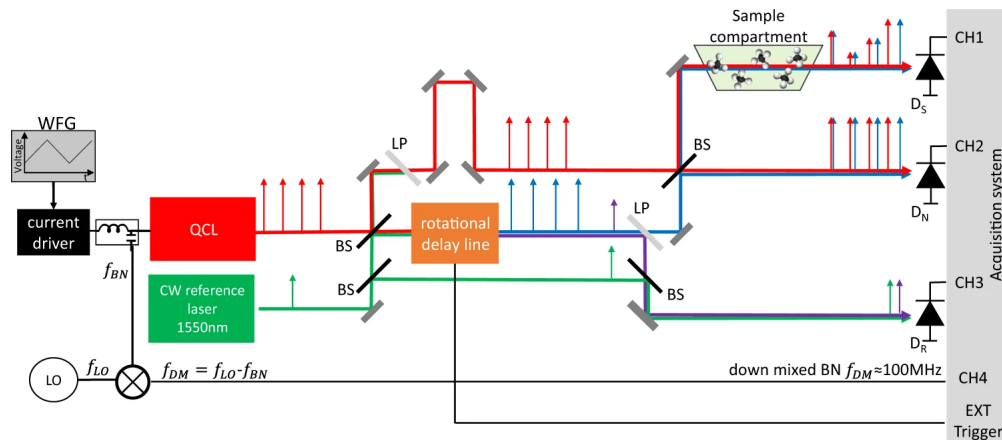


Figure 2. A CW reference laser provides a frequency reference and serves as a tool for mapping out and removing the induced frequency chirp imposed by the nonlinear rotational delay line. QCL and CW laser beam are spatially superimposed via a beam splitter (BS) and also separated via the optical low-pass filter (LP). Interferograms of the QCL are acquired with the sample (DS) and normalization (DN) detector, where the reference interferogram of the CW laser is recorded with detector DR. All measurements are synchronized with the trigger. The function generator (WFG) can be used for continuously tuning the QCL operation point by applying a ramp to the current driver of the QCL.¹

METHOD

Researchers from the Institute for Quantum Electronics in Zürich, Switzerland have developed a FT spectrometer based on a rotational motion of the scanning mirror that has the ability to perform Mid-IR dual-comb spectroscopy with a single comb source. The setup of the system can be seen in **Figure 2**. The QCL light source emits around 8 μm wavelength with a maximum emission power of 60 mW at -20°C . The emitted light is a frequency modulated comb, resulting in a linearly chirped instantaneous frequency with nearly constant intensity.¹ The centerpiece of this design is

the rotational delay line (RDL) that enhances the temporal resolution with a three-dimensional rotational retro-reflecting octagrammic prism and a static retro-reflecting system in **Figure 3**. This makes the temporal resolution proportional to the angular velocity of the rotating mirror and can reach millisecond acquisition speeds. As the number of retro-reflectors depends on a number of variables, eight reflectors are chosen for an acquisition time recording a single interferogram in around 1 ms or faster. Fortunately, the phase delay is independent of beam diameter, widening the spectroscopy applications for this system.

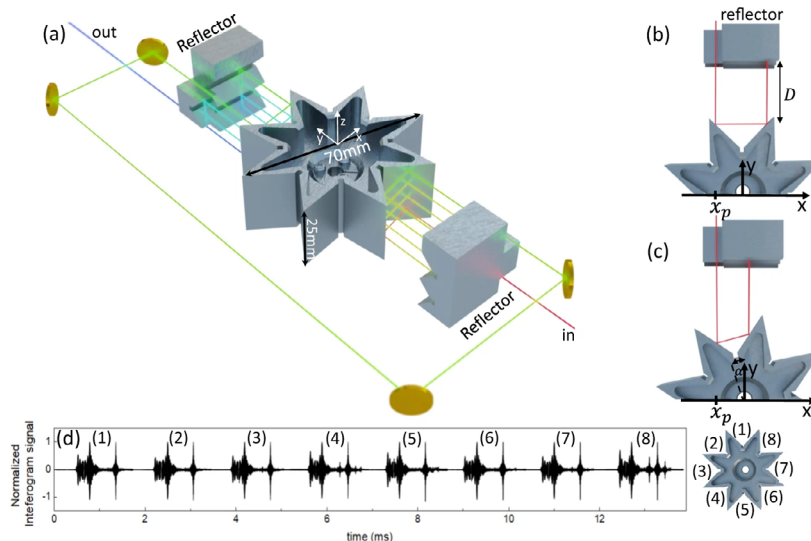


Figure 3. (a) Realization of the rotational delay line (RDL) consisting of three-dimensional rotational retro-reflecting (RR) octagrammic prism and a static retro-reflecting system (SR). The SR is arranged in such a way with respect to RR, that multiple reflections between SR and RR are possible. By exploiting the point symmetry of the system, the optical path delay can be doubled with two SR on opposite sides of the RR. (b), (c) Top view of a beam propagation in the RDL at rotation angle 0° and α . (d) Recorded interferogram on each rotational retro-reflector of the delay line.¹

To perform absorption with the rotational FT spectrometer, a background spectrum and a sample spectrum are necessary. This ensures that any background noise and light scattering is verified to be present and removed from the QCL spectrum. Before showing the high resolution of this design, further amplitude noise tests are needed. The sensitivity, signal-to-noise ratio (SNR), and stability are determined. Amplitude fluctuations were shown to be less than 1% with excellent stability over several minutes. $1/f$ noise in the system is minimized, and the system background is stabilized with deviations below 1.5% allowing for long-term measurements.

In contrast to traditional dual-comb spectroscopy with two synchronized current ramps of different slope, a single triangular current modulation ramp is applied to the QCL using Wavelength Electronics' driver. This is set to enable the overall QCL spectrum to be tuned by one frequency repetition.¹

Two demonstrations of the effectiveness of this spectrometer were performed on a 500 μm thick silicon etalon as well as on a gas chamber filled with methane. These showed the high spectral resolution as well as the fast temporal resolution of the overall design.

RESULTS

The interleaved data, of 20 consecutive interferograms that are coherently co-added before the spectrum is computed, is shown in **Figure 4(a,b)** for the silicon etalon. As is shown with the red line in the plots, the recorded interleaved spectra fit well with the computed etalon spectrum. Within a short time of 7 seconds, etalon features were resolved with resolution less than 0.15 cm^{-1} . This proves the potential of this spectrometer for sub-GHz spectroscopy.¹

The other demonstration with methane proves the more challenging conditions. Methane, at low pressures, have sub-GHz linewidths. For this sample, the QCL ramp period was increased to 25 seconds, generating thousands of interferograms that are recorded on millisecond time scales. With the interleaving process, 10 consecutive interferograms are co-added before computing each spectra.¹ The methane spectrum is shown in **Figure 4(c,d)** with sub-GHz resolved absorption lines. The zoomed in portion shows that all sub-GHz methane absorption lines can be well resolved. This is a clear advantage over the traditional FT spectrometer. This demonstrates millisecond acquisition times with 0.5 cm^{-1} (15GHz) spectral resolution, a feat impossible to achieve with rapid scan commercial FT spectrometers.¹

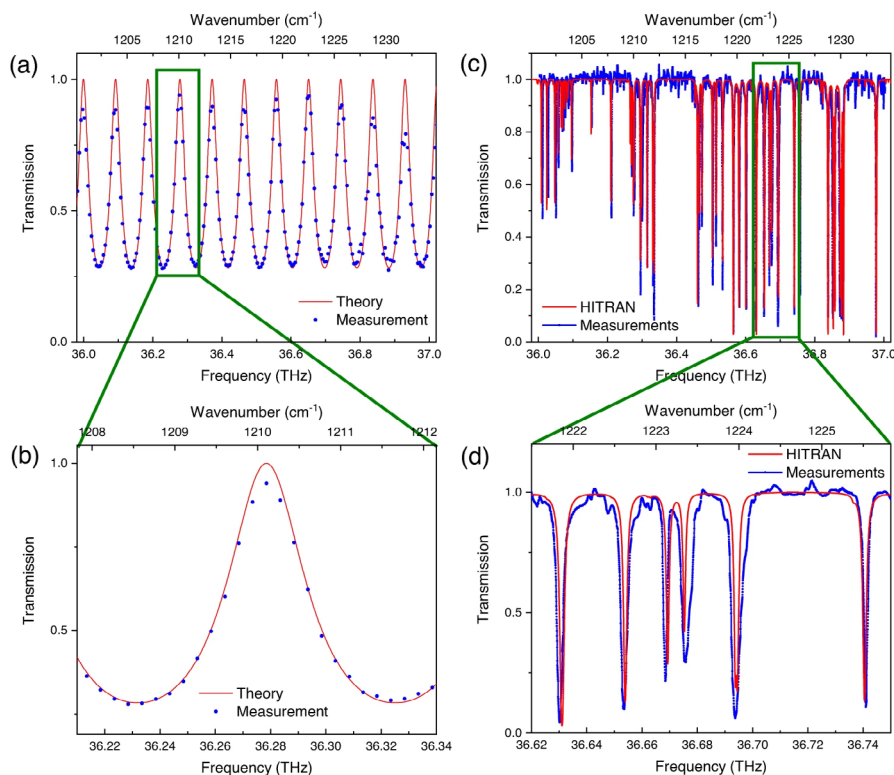


Figure 4. (a) Measured interleaving spectrum (blue dots, error bars smaller than the dots) of $\approx 500\text{ }\mu\text{m}$ thick silicon etalon within 7s with a binned frequency resolution down to 5GHz. Theoretical computed etalon transmission spectrum (red line). (b) Zoom into (a). (c) Doppler broadened methane (CH₄) spectrum recorded via interleaving (blue dots) at a pressure of 200 mbar within 25s with frequency binned resolution down to 250 MHz and the HITRAN data base reference (red line). (d) Zoom into (c) with sub-GHz resolved methane absorption lines.¹

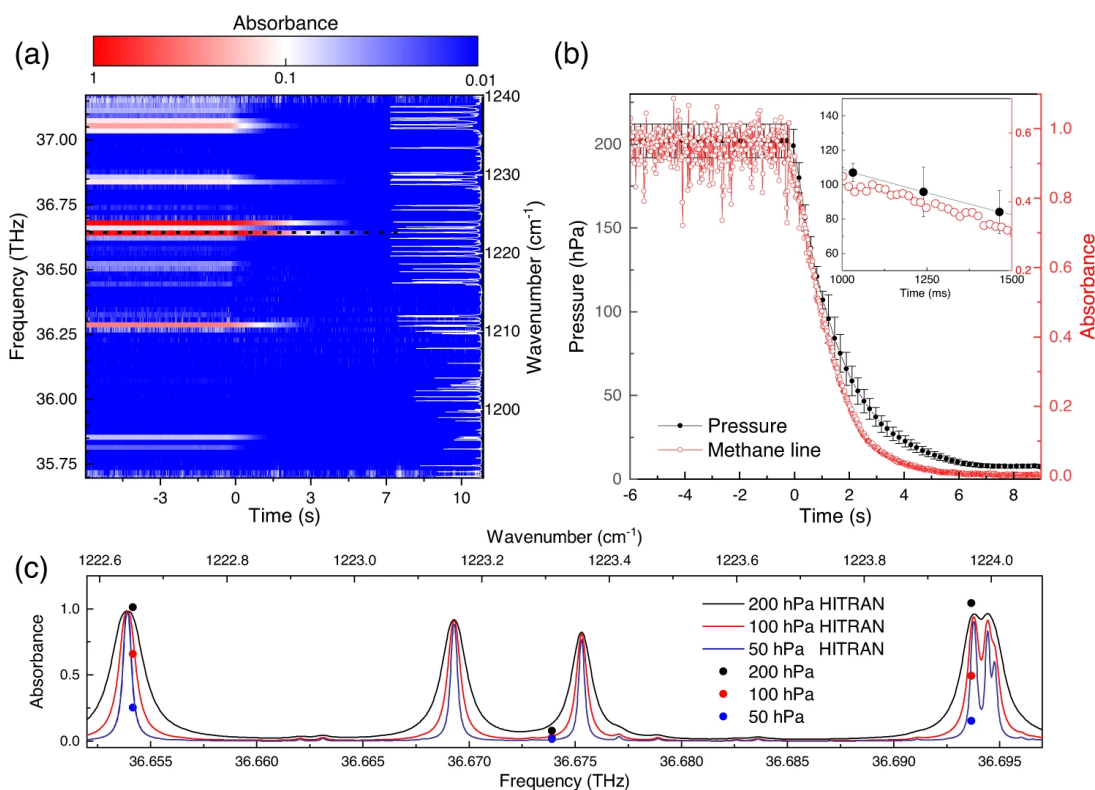


Figure 5. (a) Absorbance is computed from time resolved-transmission measurements on the filled gas cell to a pressure of 200 hPa which is evacuated via a pump. The computed methane absorbance for a pressure of 200 hPa is shown on the right side with white lines. (b) Visualization of the temporal evolution of the strongest absorption line (marked with a dotted black line) in (a) and the corresponding pressure of the gas cell (error bars are pressure gauge error). A zoom in is shown in the top right corner with 500 ms time duration starting 1 s after opening the valve. (c) Computed HITRAN absorbance of methane at a pressure of 200 hPa, 100 hPa and 50 hPa and the measured absorbance close to the strongest absorbance peak at these pressures and hence corresponding laboratory time.¹

To demonstrate the higher acquisition speeds of the developed spectrometer, time-resolved spectroscopy was performed on methane gas. Several low pressure methane absorption lines are measured as seen in **Figure 5**. There is significant agreement of spectral and temporal resolved methane absorption lines. Because each line can be seen as an independent barometer, each line can be calibrated, increasing the accuracy of the measurement.

This proof-of-principle rotational FT spectrometer enables high spectral resolution alongside fast acquisition times. Because of the ms speeds with the dual-comb spectroscopy, performed with only a single QCL frequency comb, the $1/f$ noise was significantly reduced. With the silicon etalon and the low pressure methane, high resolution was demonstrated with the interleaving technique. This could prove useful for photo-acoustic dual-comb spectroscopy or multidimensional FT spectroscopy as well.¹

With the research shown here, the majority of spectroscopy applications can benefit, ranging from reaction monitoring, leakage monitoring, and in-line process analytical technology.¹ The advantages of dual-comb and FT spectroscopy are combined and displayed for an improved QCL frequency comb as the light source. This design creates more opportunities where high speed, large optical bandwidth, and high spectral resolution are desired.

WAVELENGTH'S ROLE

Combining FT spectroscopy with dual-comb spectroscopy for fast acquisition and high-spectral resolution requires high precision and stable control of the quantum cascade laser. Wavelength Electronics' QCL driver, QCL1000 OEM, enabled precise current control with minimal electronic noise from the QCL. The driver also allows analog modulation of up to 2-3 MHz for wavelength modulation. This enabled laser tuning by changing the drive current to the QCL with a triangular current modulation ramp. As laser linewidth is a major concern for QCLs, the QCL1000 OEM minimizes noise to as low as 0.7 μA up to 100kHz as well as keeping the average current noise density to as low as 2 nA / $\sqrt{\text{Hz}}$.

The stability and low noise of the QCL supply current is critical for consistent electrical bandwidth of the QCL. Wavelengths' QCL1000 OEM, can precisely deliver up to 1 A to the laser. Additional features, such as the brown-out, over- and reverse-voltage, soft-clamping current limit, and over-temperature circuits protect the user and the QCL from electrical faults and potential damage.

The QCL1000 OEM QCL driver enables sub-second spectroscopy with high spectral resolution with low noise and stable laser output. This makes the developed rotational FT spectrometer system a reliable tool for dual-comb spectroscopy for applications in reaction and leakage monitoring and in-line process analytical technology.¹

REFERENCES

1. Markmann, S., Franckie, M., Bertrand, M., Manzoni, C., & Cerullo, G. "Frequency Chirped Fourier-Transform Spectroscopy" *Communications Physics*, **6**, 53 (2023). <https://doi.org/10.1038/s42005-023-01157-5>

USEFUL LINKS

- QCL1000 OEM [Product Page](#)

PERMISSIONS

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PRODUCTS USED

QCL1000 OEM

KEYWORDS

Fourier-transform spectroscopy, frequency chirped, spectroscopy, high spectral resolution, mid-infrared, dual-comb, quantum cascade laser, QCL, FT, laser driver, QCL1000 OEM

REVISION HISTORY

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REVISION	DATE	NOTES
A	May 2023	Initial Release