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Ultra-fast gas spectroscopy with a dual-comb spectrometer

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Summary:
Dual-comb spectroscopy has the potential to measure broadband transmission spectra fast and precisely. To employ these features for gas analysis we present a fiber-based dual-comb-spectrometer and compare the spectrum of an HCN filled cell with a simulation using the HITRAN database. Although point-to-point fluctuations across the spectrum show a rms of 4 % fit results indicate that absorptions features with (α*L) = 0.01 can be detected within measurement time of 0.5 ms.

Keywords: dual-comb-spectroscopy, gas analysis, fast acquisition, hydrogen cyanide, absorption spectroscopy

Motivation

The study of dynamics in chemical reactions or the tracking of gas concentrations is a field that profits from optical measurement techniques. With the ability to record absorption spectra with a high rate one could investigate reactions as they occur during the removal of methane - an important greenhouse gas - from the atmosphere [1]. Alternatively, the development of combustion engines or monitoring plasma compositions in industrial processes could profit from fast spectra acquisition as well.

Dual-Comb spectroscopy

A frequency comb can be described as a pulsed laser with its optical spectrum composed of multiple discrete modes with identical spacing f_r. Their use as a ruler in the optical domain allows for extraordinary precision in frequency metrology [2]. As the comb spectrum usually covers a broad spectral range, the attenuation caused by a probe, here gaseous samples, can be determined. By superimposing the probing comb with a reference comb - often referred to as dual-comb-spectroscopy [3] - beatings between pairs of comb modes are generated. This effectively realizes a multi-heterodyne detection scheme resulting in a frequency-comb in the radio frequency regime. Here the spacing of the rf-combs Δf_r equals the difference in the comb mode spacing of the respective combs. Interestingly the acquisition rate is also Δf_r. Knowing f_r and Δf_r one can directly map the RF-comb to the optical domain and retrieve the transmission spectra of the probe with an optical resolution f_r - here in the order of hundreds of MHz. However, using two combs generated separately as with Titan Sapphire- or fiber-lasers requires a significant effort to ensure phase-synchronization.

Experimental setup

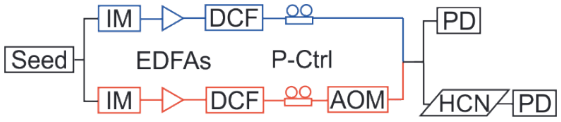

Fig. 1. Sketch of experimental setup for a fiber-based dual-comb-generator. The cw-laser (Seed) is split into two branches (red/blue). Using intensity modulation (IM), amplification (EDFAs) and spectral broadening in a dispersion compensating fiber (DCF) two combs are generated. They are polarization-controlled (P-Ctrl). One comb is shifted in frequency by an acousto-optic modulator (AOM). After superposition a part the dual-comb signal is fed to a photodetector (PD) as reference while the other part passes a sealed cell filled with hydrogen cyanide (HCN) before being detected.

Following a different approach [4] we generate two frequency combs using a common continuous wave (cw) fiber-laser (emitting at 1550 nm, frequency drift below 10 MHz/min) leading to mutual-coherence of the combs (see Fig. 1). For each comb the cw-laser is intensity modulated by electro-optic modulators resulting in a pulse train with f_r = 275 MHz repetition rate (f_r +10 kHz for the second comb). The spectrum now consists of few 100 modes. After amplification the pulse trains are launched into a dispersion compensating fiber for spectral broadening [5] resulting in 1800 comb modes spanning an optical

bandwidth of 490 GHz. Before superimposing both combs their polarizations are aligned and one comb is shifted by 25 MHz in frequency with an acousto-optic modulator to allow for mapping of the radio-frequency comb to the optical domain without ambiguity. We use two detection channels where one serves as reference. In the probe channel the dual-comb signal passes a multi-pass cell with 78 cm length filled with hydrogen cyanide at 100 Torr. For both channels the multi-heterodyne signal is recorded by a fast photodetector connected to a digitizer. The power spectrum reveals the radio-frequency combs. To remove the comb envelope structure, the probe channel data is divided by the reference channel data. In addition, a constant scaling factor is applied to account for different optical powers reaching the detectors (see Fig. 2).

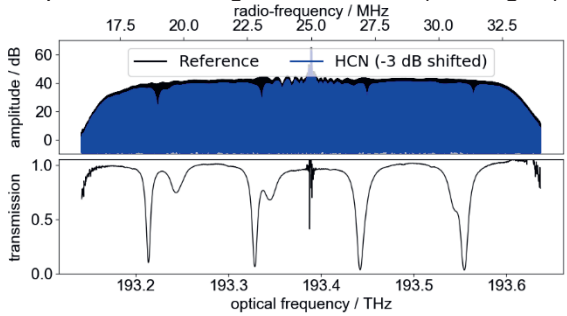

Fig. 2. (top) Dual-comb spectra in the radio-frequency domain showing absorptions due to hydrogen cyanide (blue) compared to a reference channel (black). (bottom) Transmission spectrum mapped to the optical regime.

Results

With this configuration we typically observe 1800 comb modes with amplitudes 42 dB above the noise floor in average for 1 s measurement time. This corresponds to a figure of merit [3] of 2.7 x 10^7 $Hz^{1/2}$. A transmission spectrum measured within 0.5 ms is compared with a simulation based on the HITRAN database [6] (see Fig. 3). While the 0.5-ms-data shows a noise rms of 4% the fit allows one to detect absorption features with (α*L) = 0.01 at this short time scale.

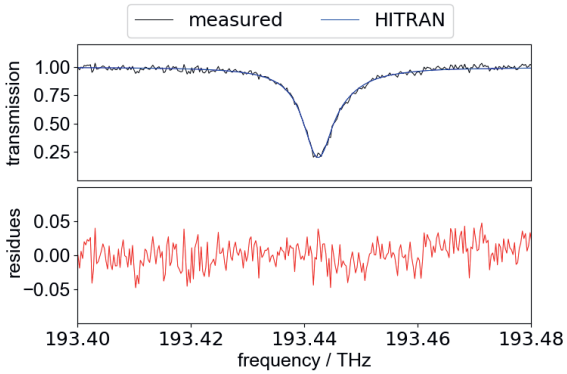

Fig. 3. (top) Zoom-in on HCN absorption feature at 193.44 THz (measurement and HITRAN data).(absorption path 78 cm, prefilled with hydrogen cyanide at 100 Torr. (bottom) Residues show the impact of point-to-point fluctuations across the spectrum.

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