High-Resolution and Gapless Dual Comb Spectroscopy with Current-Tuned Quantum Cascade Lasers

Michele Gianella,[1] Akshay Nataraj,[1] Béla Tuzson,[1] Jérôme Faist,[2] and Lukas Emmenegger[1]
[1]*Laboratory for Air Pollution & Environmental Technology, Empa, Überlandstrasse 129, 8600 Dübendorf, Switzerland*
[2]*Institute for Quantum Electronics, ETH Zurich, Auguste-Piccard-Hof 1, 8093 Zurich, Switzerland*
michele.gianella@empa.ch

Abstract: We measured gapless, high-resolution absorption spectra spanning $55\ \mathrm{cm}^{-1}$ by simultaneous current-modulation of two quantum cascade laser frequency combs. Detector noise limited spectra were obtained in as little as $10\ \mathrm{ms}$ with aresolution of a few MHz.

1. Introduction

Quantum cascade laser frequency combs (QCL-FCs) [1] are compact frequency comb sources providing direct access to the mid-infrared (MIR) spectral range, where many industrially, environmentally and medically relevant molecules possess strong and characteristic fundamental ro-vibrational absorption bands. These relatively recent laser sources are most commonly employed in the so-called *dual comb* scheme, in which a *local oscillator* comb is overlapped with the *interrogating comb* at the detector, generating a multi-heterodyne beat note signal which allows the measurement of the amplitude of the individual comb lines [2]. QCL-FCs hold much promise for spectroscopic applications requiring fast temporal resolution and broad spectral coverage that cannot be achieved with traditional sources, such as distributed feedback lasers [3]. Furthermore, even free-running QCL-FCs show sufficient mutual coherence that active locking – either mutual or to a common reference – is not necessary, which strongly reduces the complexity of the experimental setup [4].

For absorption spectroscopy in the gas phase, the spectral point spacing – given by the free spectral range (FSR) of the laser ($5-15$ GHz) – is not sufficient to resolve Doppler-broadened transitions, even though the spectral resolution – given by the comb lines' linewidth (~ 1 MHz) – is perfectly adequate. This can be overcome by interleaving sequentially acquired spectra with an incremental offset of the comb line frequencies, thereby reducing the spectral point spacing by several orders of magnitude. The product of this technique are highly resolved, gapless MIR spectra based on current-tuned quantum cascade lasers.

2. Experimental setup

A dual-comb spectrometer based on the phase-sensitive (dispersive) design with current modulation applied on both lasers is shown schematically in **Fig. 1**. The two laser sources are InGaAs/AlInAs on InP-based dual-stack QCLs [5]. The comb line frequencies are swept by applying a triangular current modulation to the lasers. To prevent the beat note frequencies from moving outside of the detection bandwidth (1 GHz), the drive currents of *both* lasers are modulated simultaneously. The interferograms acquired concurrently by the two detectors are partitioned into slices of 16.4 μs length and Fourier-transformed. In each slice, the beat notes are identified and their amplitudes are determined. With a background measurement, obtained by removing or evacuating the sample cell, the sample attenuation (absorbance) and phase-shift (refractive index) is extracted from the measured beat note amplitudes.

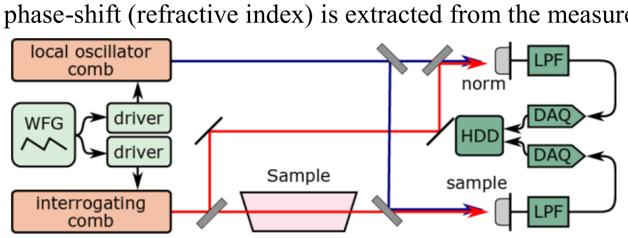

Fig. 1. Scheme of the dual comb spectrometer. WFG: waveform generator, sample/norm: photodetectors, LPF: low-pass filter, DAQ: data acquisition board, HDD: hard disk

3. Results

Figure 2 shows the absorption spectrum of the 14 cm sample cell filled with 107 hPa of methane. A linear current ramp was applied to both lasers which resulted in the tuning of the comb line frequencies by about 10 GHz (the FSR of the lasers) during 120 ms (tuning rate: 83 MHz/ms). Approximately 30,000 spectra were acquired, resulting in a frequency offset of 300 kHz between successive spectra. The spectral resolution in Fig. 2 is 30 MHz after smoothing. The modulation of the baseline is due to a pair of optical fringes resulting from the uncoated windows of the sample cell, and the areas of increased signal noise correspond to frequencies with low-power of the lasers. Faster sweeps of 10 ms duration (tuning rate: 1 GHz/ms) with a resolution of a few MHz are also possible employing high bandwidth and low noise laser current drivers. Noise analysis shows that the measurements are detector-noise limited.

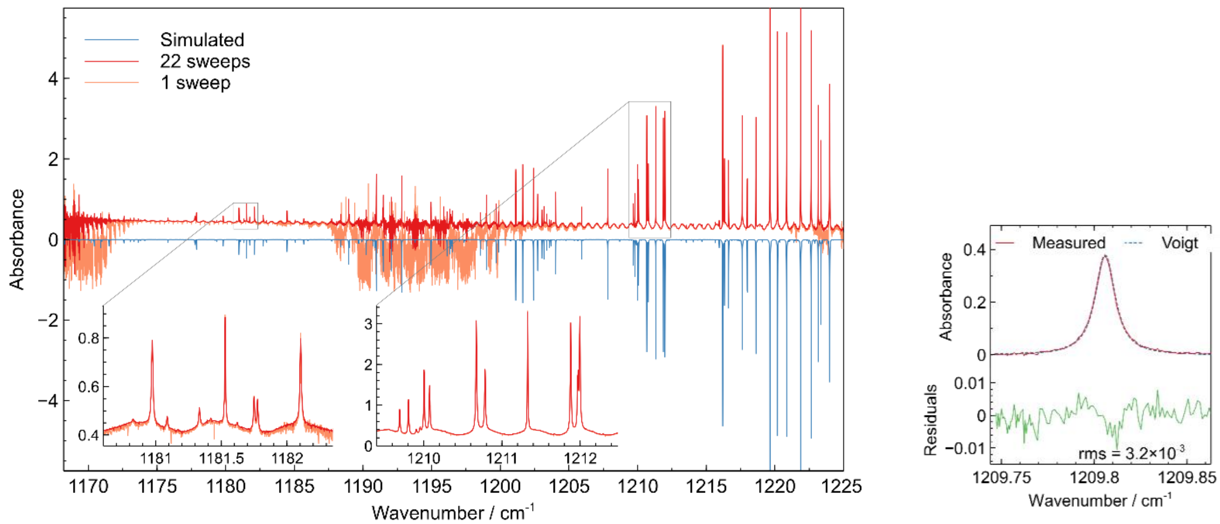

Fig. 2. *Left:* Absorption spectrum of methane. The insets show zooms of the data in the highlighted regions. *Right:* Methane transition at 1209.8 cm^{-1} with Voigt fit and residuals. Conditions: pressure: 107 hPa, path length:14 cm, spectral resolution: 0.001 cm^{-1}.

5. Conclusions and Outlook

The point spacing given by the FSR of QCL-FCs can be covered by interleaving sequentially acquired offset spectra. This is achieved through rapid (~1 GHz /ms) current modulation of the laser sources. Further improvements in terms of stability and sensitivity may be obtained through radio frequency injection [6] and the employment of an optical cell with much longer path length.

6. References

1. A. Hugi, G. Villares, S. Blaser, H. C. Liu, and J. Faist, "Mid-infrared frequency comb based on a quantum cascade laser," Nature **492**, 229–233 (2012).
2. I. Coddington, N. Newbury, and W. Swann, "Dual-comb spectroscopy," Optica **3**, 414–426 (2016).
3. J. L. Klocke, M. Mangold, P. Allmendinger, A. Hugi, M. Geiser, P. Jouy, J. Faist, and T. Kottke, "Single-Shot Sub-microsecond Mid-infrared Spectroscopy on Protein Reactions with Quantum Cascade Laser Frequency Combs," Anal. Chem. **90**, 10494–10500 (2018).
4. G. Villares, A. Hugi, S. Blaser, and J. Faist, "Dual-comb spectroscopy based on quantum-cascade-laser frequency combs," Nat. Commun. **5**, 1–3 (2014).
5. P. Jouy, J. M. Wolf, Y. Bidaux, P. Allmendinger, M. Mangold, M. Beck, and J. Faist, "Dual comb operation of λ ~ 8.2 μm quantum cascade laser frequency comb with 1 W optical power," Appl. Phys. Lett. **111**, 141102 (2017).
6. J. Hillbrand, A. M. Andrews, H. Detz, G. Strasser, and B. Schwarz, "Coherent injection locking of quantum cascade laser frequency combs," Nat. Photonics **13**, 101–104 (2019).