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High-Resolution Quantum Cascade Laser Dual-Comb Spectroscopy with Accurate Absolute Frequency Scale

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Abstract: We present a mid-infrared dual comb spectrometer featuring fast acquisition (54 ms), high spectral resolution (630 kHz), and high frequency accuracy (2 MHz), in the 7.8 um wavelength range.

1. Introduction

Quantum cascade laser (QCL) frequency combs [1] in dual-comb arrangement [2] are well suited to tasks requiring high temporal resolution [3]. High *spectral* resolution is achieved by taking advantage of their current-tunability, to sweep sweeping the interrogating comb and interleaving multiple spectra [4,5]. The absolute frequency axis can easily be determined if the repetition frequency and one comb line frequency are known at every instant of the sweep.

2. Experimental setup

The dual-comb spectrometer employs two QCL frequency combs covering the range 1265-1305 cm^{-1} (Fig. 1) with a free spectral range (or repetition frequency) of f_{rep} = 9.88 GHz. The sample is interrogated within a 36 m multi-pass cell (MPC, Aerodyne Research, AMAC-36LW). The currents of QCL1 and QCL2 are modulated with a sawtooth waveform so that the teeth of the interrogating comb shift by 10 GHz ($\approx f_{rep}$) in 54 ms. The sweep is divided into ~16,000 slices, yielding a spectral point spacing of 630 kHz. A frequency-locked, single-mode distributed feedback QCL (DFB, Alpes Lasers) acts as the optical frequency reference.

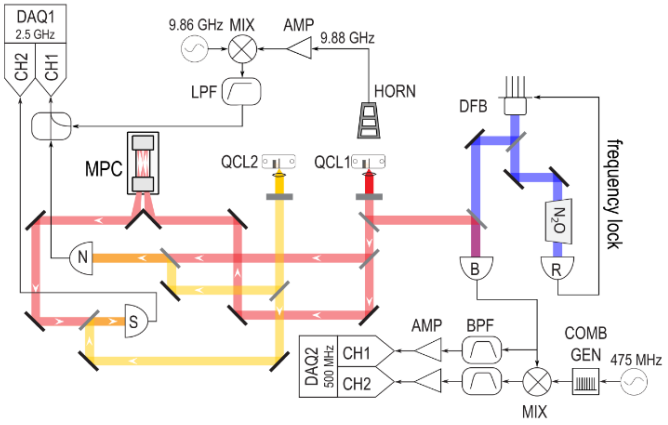

Fig. 1. Dual-comb spectrometer with frequency-stabilized DFB laser, as well as repetition frequency and comb tooth frequency measurement circuits. COMBGEN: harmonic RF comb generator; MIX: mixer; AMP: amplifier; LPF/BPF: low/band-pass filter; HORN: microwave horn antenna; N/S/B/R: photodetectors.

The DFB beam is heterodyned with the beam of QCL1 on photodetector B. The lowest beat note frequency, ν_Δ, is mixed with a harmonic RF comb with a period of 475 MHz. As QCL1 is tuned, ν_Δ chirps, passing through the harmonics of 475 MHz. The timing of each pass is detected via a band-pass filter and a logarithmic amplifier. From the recorded timing of the pulses, a time-frequency "calibration" curve is obtained for the comb tooth of QCL1. In combination with the measurement of f_{rep}, the frequencies of all comb teeth can be computed.

The intermode beat [6] of QCL1 (~9.88 GHz) is captured by a microwave horn antenna (HORN, Aaronia, PowerLOG 40400) placed close to QCL1. After down-mixing to ~20 MHz, the beat is combined with the multi-heterodyne beat signal from detector N. In processing the dual-comb data, the intermode beat appears as an additional beat note with frequency around 20 MHz from which f_{rep} is computed.

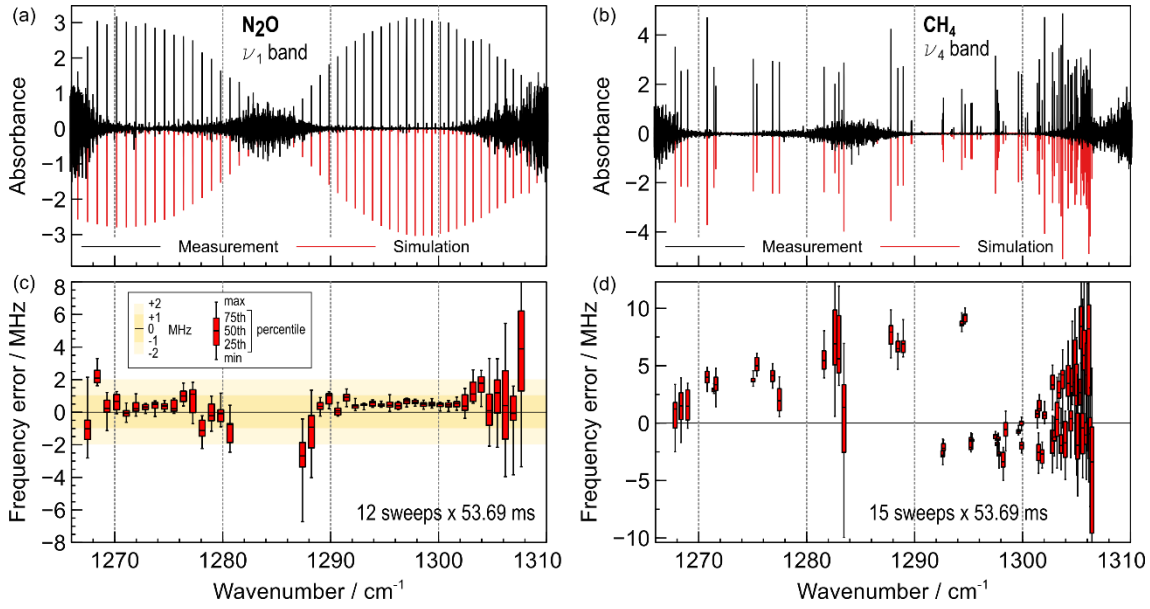

Fig. 2. Measured and computed absorption spectra of: (a) the ν_1 fundamental band of N_2O and (b) the ν_4 fundamental band of CH_4. Line centre frequency errors relative to: (c) the values in [7] and (d) the values in HITRAN2016 [8].

3. Results and discussion

We measured the ν_1 fundamental band of N_2O at 1.5 mbar in the MPC (Fig. 2a) and the ν_4 fundamental band of CH_4 at 1.8 mbar (Fig. 2b). Accurate published transition frequencies for N_2O [7,9] allow us to test the accuracy of our frequency scale. The frequency scale is accurate to within ± 1 MHz between 1269 and 1281 cm^{-1} and between 1288 and 1303 cm^{-1} (Fig. 2c). Only three of the 43 measured transitions exhibit an error greater than 2 MHz, and each of these exceptions is located in a region of poor signal-to-noise ratio (SNR). For methane, we compare the measured line centre frequencies with the HITRAN2016 database [8] (Fig. 2d). The discrepancy between our measurement and the HITRAN database is up to 10 MHz. Since it qualitatively matches a recent measurement [10], we account for it through the uncertainty of the database, given as between 30 and 300 MHz.

4. Conclusions

We have demonstrated dual-comb spectroscopy in 54 ms with a resolution of 630 kHz and with frequency accuracy better than 2 MHz over almost the entire spectral coverage of the comb source.

5. 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] M. Gianella, A. Nataraj, B. Tuzson, P. Jouy, F. Kapsalidis, M. Beck, M. Mangold, A. Hugi, J. Faist, and L. Emmenegger, "High-resolution and gapless dual comb spectroscopy with current-tuned quantum cascade lasers," Opt. Express **28**, 6197–6208 (2020).
[5] M. Gianella, S. Vogel, V. J. Wittwer, T. Südmeyer, J. Faist, and L. Emmenegger, "Frequency axis for swept dual-comb spectroscopy with quantum cascade lasers," Opt. Lett. **accepted**, (2022).
[6] M. Piccardo, D. Kazakov, N. A. Rubin, P. Chevalier, Y. Wang, F. Xie, K. Lascola, A. Belyanin, and F. Capasso, "Time-dependent population inversion gratings in laser frequency combs," Optica **5**, 475–478 (2018).
[7] A. Hjältén, M. Germann, K. Krzempek, A. Hudzikowski, A. Gluszek, D. Tomaszewska, G. Sobon, and A. Foltynowicz, "Optical frequency comb Fourier transform spectroscopy of (N2O)-N-14-O-16 at 7.8 µm," J. Quant. Spectrosc. Radiat. Transf. **271**, 107734 (2021).
[8] I. E. Gordon *et al.*, "The HITRAN2016 molecular spectroscopic database," J. Quant. Spectrosc. Radiat. Transf. **203**, 3–69 (2017).
[9] B. AlSaif, M. Lamperti, D. Gatti, P. Laporta, M. Fermann, A. Farooq, O. Lyulin, A. Campargue, and M. Marangoni, "High accuracy line positions of the v1 fundamental band of 14N216O," J. Quant. Spectrosc. Radiat. Transf. **211**, 172–178 (2018).
[10] A. Hjältén, M. Germann, I. Sadiek, C. Lu, F. S. Vieira, K. Krzempek, A. Hudzikowski, A. Głuszek, D. Tomaszewska, M. Stuhr, G. Soboń, and A. Foltynowicz, "Fourier transform spectroscopy using difference frequency generation comb sources at 3.3 µm and 7.8 µm," in OSA Optical Sensors and Sensing Congress (Optical Society of America, 2021), p. JTu4D.3.