

Frequency Scale Calibration for High-Resolution Quantum Cascade Laser Dual-Comb Spectroscopy

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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 μm wavelength range. © 2022 The Author(s)

1. Introduction

Optical frequency combs are coherent sources for infrared absorption spectroscopy that promise broad spectral coverage with very high spectral and temporal resolution. The repetition frequency of an optical frequency comb is governed by the length of the laser resonator and determines the point spacing of the measured spectrum. To decrease the point spacing, multiple spectra with small increments or decrements of the repetition or carrier-envelope offset frequencies are interleaved [1]. To interleave the spectra correctly, both the repetition frequency and the carrier-envelope offset frequency must be known for every spectrum.

2. Experimental setup

We employ a dual-comb spectrometer with two QCL frequency combs covering the range 1265-1305 cm^{-1} with a repetition frequency of $f_{\text{rep}} = 9.88 \text{ GHz}$ (Fig. 1a). The interrogated samples is in 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 ($\approx f_{\text{rep}}$) in 54 ms. The sweep is divided into $\sim 16,000$ slices which, after appropriate interleaving, yield a composite spectrum with a spectral point spacing of 630 kHz. The optical reference frequency is provided by a frequency-locked (to a molecular transition), single-mode distributed feedback QCL (DFB, Alpes Lasers).

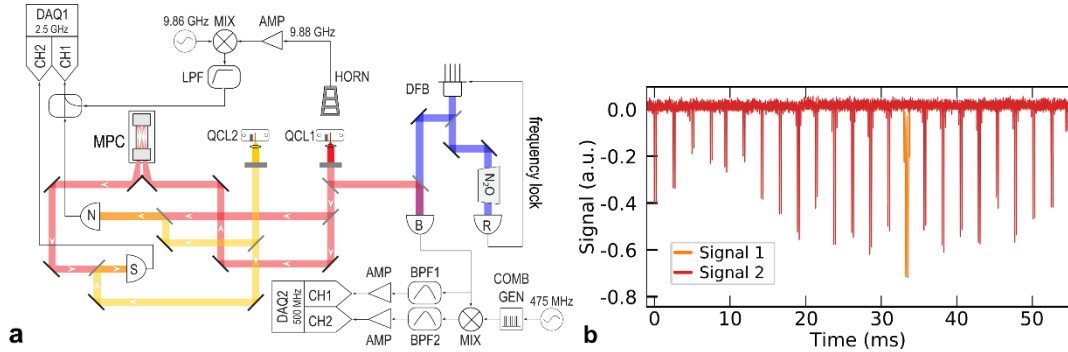


Fig. 1. (a) Dual-comb spectrometer with frequency-stabilized DFB laser, as well as repetition frequency and comb tooth frequency measurement circuits. COMBGEN: harmonic RF comb generator; MIX: mixer; AMP: amplifier; LPF/BPF1,2: low/band-pass filter; HORN: microwave horn antenna; N/S/B/R: photodetectors. (b) Signals recorded on channels 1 and 2 of DAQ2 during sweeping.

The DFB beam is heterodyned with the beam of QCL1 on photodetector B. The lowest beat note frequency, $\nu_{\Delta}(t) = \nu_0(t) - \nu_{\text{DFB}}$, generated by the beating of the comb tooth with frequency $\nu_0(t)$ with the DFB laser, ν_{DFB} , is mixed with a synthesized harmonic RF comb with a period of 475 MHz. At the two instants, t_{*}^{\pm} , where $\nu_{\Delta}(t_{*}^{\pm}) = \pm CF$, ($CF \approx 30 \text{ MHz}$ is the centre frequency of BPF1 and BPF2) the bandpass filter BPF1 transmits two pulses (signal 1 in Fig. 1b). We then assume that $\nu_{\Delta}(\bar{t}_{*}) = 0 = \nu_0(\bar{t}_{*}) - \nu_{\text{DFB}}$, where $\bar{t}_{*} = (t_{*}^{+} + t_{*}^{-})/2$. We now know that $\nu_0(\bar{t}_{*}) = \nu_{\text{DFB}}$. Similarly, at all time instants, t_n^{\pm} ($n = \pm 1, \pm 2, \dots$), where $\nu_{\Delta}(t_n^{\pm}) = n \cdot 475 \text{ MHz} \pm CF$, the bandpass filter BPF2 transmits a pulse (signal 2 in Fig. 1b). Following the same reasoning as before, we find that $\nu_0(\bar{t}_n) = \nu_{\text{DFB}} + n \cdot 475 \text{ MHz}$ (where $\bar{t}_n = (t_n^{+} + t_n^{-})/2$). Hence, the teeth of the synthetic 475 MHz comb act as markers, each providing a double pulse centred at \bar{t}_n ($n = \pm 1, \pm 2, \dots$) at which time the comb tooth has frequency

$\nu_0(\bar{t}_n) = \nu_{\text{DFB}} + n \cdot 475 \text{ MHz}$. From the recorded times, \bar{t}_n , of the pulses, a time-frequency "calibration" curve is obtained for $\nu_0(t)$.

The intermode beat [2] of QCL1 ($\sim 9.88 \text{ GHz}$) is captured by a microwave horn antenna (HORN, Aaronia, PowerLOG 40400) placed close to QCL1. After down-mixing to $\sim 20 \text{ MHz}$, the beat is combined with the multi-heterodyne beat signal from detector N. It thus appears as an additional beat note with frequency around 20 MHz from which f_{rep} is computed.

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 [1,3] allow us to test the accuracy of our frequency scale. The frequency scale is accurate to within $\pm 1 \text{ MHz}$ 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 [4] (Fig. 2d). The discrepancy between our measurement and the HITRAN database is up to 10 MHz . Since it qualitatively matches a recent measurement [5], we account for it through the uncertainty of the database, given as between 30 and 300 MHz .

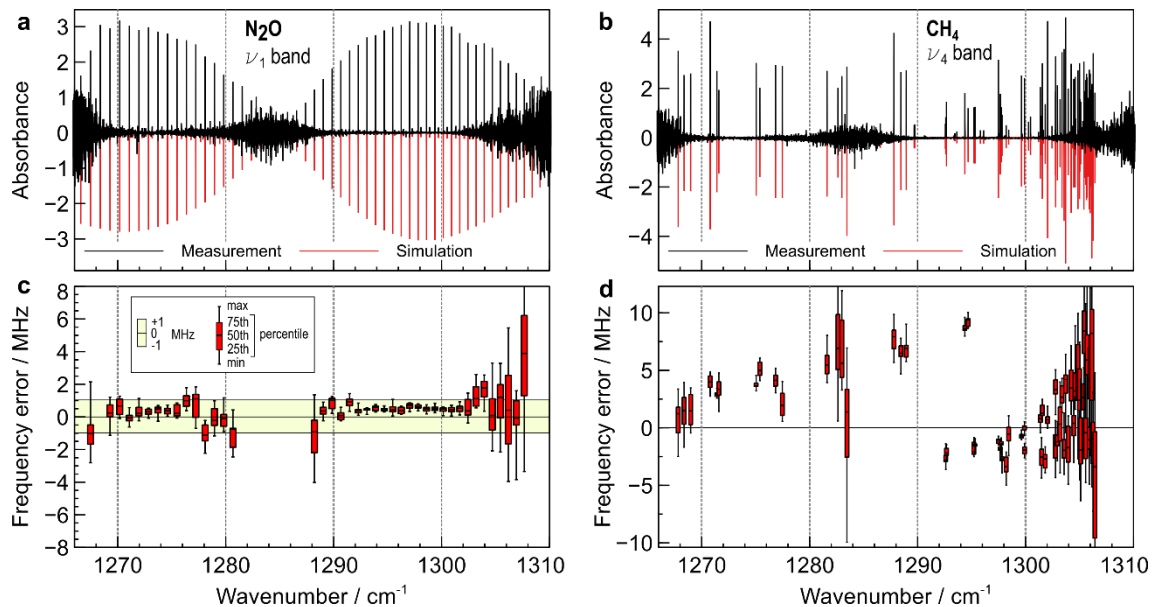


Fig. 2. Measured and computed absorption spectra of: (a) the ν_1 fundamental band of N_2O and (b) the ν_4 fundamental band of CH_4 . Line centre frequency errors relative to: (c) the values in [1] and (d) the values in HITRAN2016 [4].

4. Conclusions

We have demonstrated interleaved dual-comb spectroscopy in 54 ms with a resolution of 630 kHz and with frequency accuracy better than 2 MHz over nearly 40 cm^{-1} .

5. References

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