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

M. Gianella[1], M. Singleton[1,*], K. N. Komagata[2], S. Vogel[1], M. Bertrand[3], J. Faist[3], T. Südmeyer[2], and L. Emmenegger[1]

1 Laboratory for Air Pollution / Environmental Technology, Empa, Überlandstrasse 129, 8600 Dübendorf, Switzerland
2 Laboratoire Temps-Fréquence, Université de Neuchâtel, Avenue de Bellevaux 51, 2002 Neuchâtel, Switzerland
3 Institute for Quantum Electronics, ETH Zurich, Auguste-Piccard-Hof 1, 8093 Zurich, Switzerland

QCL based frequency combs (FCs) are bright, broadband mid-infrared sources [1]. While their repetition rates are typically ~10 GHz, laser linewidth limited resolution (MHz) is possible through spectral interleaving [2]. Combining synchronized tuning of the two FC lasers with fast acquisition and signal processing algorithms, we demonstrated real-time spectral retrieval (duty cycle ~50% at a 100 ms pull rate) and significant data volume reduction for long-term continuous monitoring [2].

An absolute RF ↔ optical frequency map requires knowledge of at least two comb parameters (f_{rep}, and either f_{ceo}, or any comb line frequency). The repetition rate can be measured directly from the bias line in QCLs, but the offset frequency is generally unknown. Either an auxiliary measurement needs to be taken, e.g. with a calibrated, unbalanced Mach-Zehnder interferometer [3], or the frequency axis can be reconstructed relying on several narrow, well-known absorption lines [2].

In a new approach, we perform an absolutely referenced absorption spectroscopy of the ν_1 band of N_2O. For this, one tooth of the interrogating comb at $\nu_x(t)$ is heterodyned against a DFB-QCL locked to a known transition of N_2O. A beat at $f(t)= \nu_x(t)-\nu_{DFB}$ results, which chirps with the modulation. This is downmixed against an RF comb f_q = q x 475 MHz, and sent through a bandpass filter at 20 MHz such that signal appears twice per crossing, when $|f(t)-f_q|$ = 20 MHz is fulfilled. The true intersection $f(t=T_q)=f_q$ lies between those events. This allows us to construct an absolute frequency map for $\nu_x(t)$ $[T_0, T_1, \ldots] \rightarrow [\nu_{DFB}, \nu_{DFB} +475 \text{ MHz}, \ldots]$, and by extension the whole comb by knowledge of $f_{rep}(t)$. Comparing to data in [4], we find for a 54 ms acquisition an accuracy better than 2 MHz.

We are now adopting the approach for a spectrometer geared towards real-time, in-situ monitoring for non-ablative femtosecond laser machining. Phase changes (amorphous ↔ crystalline) are induced, accompanied by spectral signatures in the mid-IR, yielding information on the quality of the process.

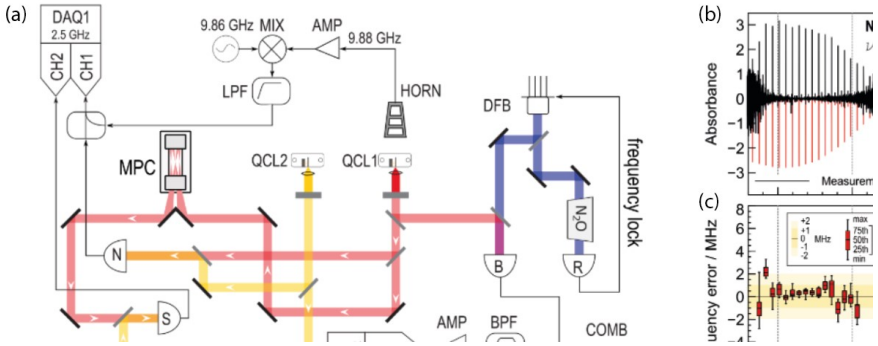

Figure 1: (a) Absolutely referenced dual-comb spectroscopic setup. (b) absorption spectra of the ν_1 fundamental band of N_2O (c) frequency error compared to the literature data [4]

[1] A. Hugi, G. Villares, S. Blaser, et al. *Nature* **492**, 229–233 (2012).
[2] M. Gianella, A. Nataraj, B. Tuzson, et al, *Opt. Express* **28**, 6197–6208 (2020).
[3] M. Gianella., S. Vogel, V. J. Wittwer, et al. *Opt. Letters*, **47(3),** 625-628. (2022).
[4] A. Hjältén, M. Germann, K. Krzempek, et al., *J. Quant. Spectrosc. Radiat. Transf.* **271**, 107734 (2021)