Optica High-brightness Sources and Light-driven Interactions Congress 2022 (EUVXRAY, HILAS, MICS) © Optica 2022

Comb-Calibrated Spectroscopy using a Quantum Cascade Laser Frequency Comb in the Long-Wave Infrared

K. N. Komagata,[1,*] M. Gianella,[2] P. Jouy,[3] F. Kapsalidis,[4] M. Shahmohammadi,[4] M. Beck,[4] R. Matthey,[1] V. J. Wittwer,[1] A. Hugi,[3] J. Faist,[4] L. Emmenegger,[2] T. Südmeyer,[1] and S. Schilt[1]

[1] *Laboratoire Temps-Fréquence, Institut de Physique, Université de Neuchâtel, 2000 Neuchâtel, Switzerland*
[2] *Laboratory for Air Pollution / Environmental Technology, Empa, 8600 Dübendorf, Switzerland*
[3] *IRsweep AG, Laubisrütistrasse 44, 8712 Stäfa, Switzerland*
[4] *Institute for Quantum Electronics, ETH Zurich, 8093 Zurich, Switzerland*
**kenichi.komagata@unine.ch*

Abstract: We demonstrate that mid-infrared quantum cascade laser frequency combs are highly suitable as high-accuracy frequency references. We exploit a fully-stabilized quantum cascade laser frequency comb to perform spectroscopy with 100-kHz frequency accuracy at 7.7 µm.

1. Introduction

Molecular transition frequencies can be measured with the highest accuracy using referenced optical frequency combs (OFCs). In this regard, the mid-infrared (MIR) region is of particular interest, as many molecules exhibit strong and characteristic fundamental absorption bands in this spectral range. However, it is challenging to perform frequency-calibrated spectroscopy in the MIR, as established referenced OFCs sources lie in the near-infrared (NIR). Typically, nonlinear frequency conversion is required to either up-convert a single-frequency MIR lasers for referencing to a NIR OFC [1], or down-convert the NIR OFC into the MIR spectral range [2]. Alternatively, quantum cascade laser (QCL) OFCs could be used, which directly emit in the MIR [3]. This chip-based solution circumvents the need for complex nonlinear frequency conversion.

In this work, we present an absolute referencing scheme exploiting a QCL-OFC. We lock one comb line to a known molecular transition and stabilize the comb-line spacing via electrical radio-frequency (RF) injection locking. The frequency stability of our setup was previously characterized [4]. Here, we use the QCL-OFC to perform comb-calibrated spectroscopy with high frequency accuracy in the 7.7 µm wavelength range.

2. Experimental setup

We implemented the frequency-referenced QCL-OFC as shown in Fig. 1(a), similarly to our previous work [4]. We locked a distributed-feedback (DFB) QCL labelled DFB-QCL1 to the R(8) molecular transition in the ν_1 fundamental band of N_2O at about 1292.3 cm^{-1} using wavelength modulation spectroscopy (WMS). A fast modulation at a frequency f_{wms} of about 50 MHz was applied to the laser current via a custom-made bias-tee (BT). After passing through a gas cell filled with N_2O at 5-mbar pressure, the beam was detected using a photodetector (PD2).

The detector DC signal provided the optical transmission through the gas cell whereas the AC signal was demodulated at frequency f_{wms} to generate a WMS signal featuring a slope of about 25 mV/MHz [see Fig. 1(b,c)]. The error signal was fed to a proportional-integrator (PI) servo-controller to generate a feedback signal to stabilize the DFB-QCL1 emission frequency to the zero-crossing point via current modulation. The absolute frequency of the DFB-QCL1 was assessed from the offset between the zero-crossing point of the error signal and the transition center-frequency, determined using a Voigt fit of the cell transmission.

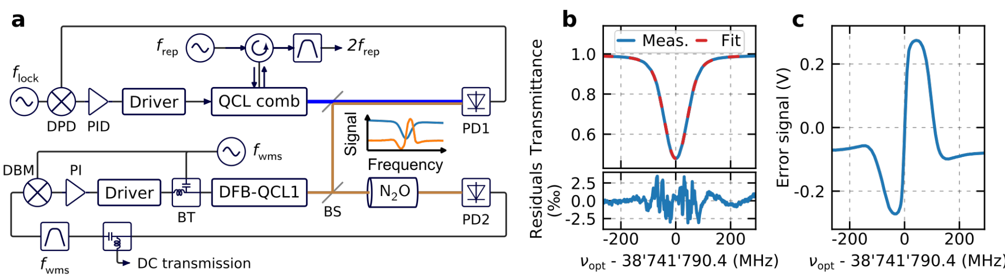

Fig. 1. Stabilization of the QCL-OFC. (a) Setup to reference the QCL comb to a molecular transition. DPD: digital phase detector, BS: beamsplitter, DBM: double balanced mixer. (b) Transmission through the gas cell and Voigt fit. (c) Error signal.

The QCL-comb chip was designed and fabricated at ETH Zurich. A wire bond connects the laser active area near the back facet to a dedicated (RF) port on a printed-circuit board (PCB) designed by IRsweep AG, which allows for efficient RF injection and extraction of the intermode beat signal of frequency f_{rep}. The intermode beat frequency was stabilized by RF injection-locking at 9.75 GHz using 20 dBm of RF power. A circulator was used to simultaneously inject the master signal and extract the second harmonic of the intermode beat for monitoring.

A part of the DFB-QCL1 beam was heterodyned with the QCL-OFC on a fast photodetector (PD1). The heterodyne beat was compared in phase with a reference signal of frequency f_{lock}. The resulting phase error signal was fed to a P-I^2-D servo-controller acting on the QCL-OFC driving current, establishing a phase lock between one comb line of the QCL-OFC and DFB-QCL1. Hence, the frequency of all comb lines is stabilized and known.

Next, we phase-locked a second laser labelled DFB-QCL2 to another comb line, approximately 500 GHz away from DFB-QCL1, by heterodyning DFB-QCL2 and the QCL-OFC on a fast photodetector [Fig. 2(a)]. Their beat signal was mixed with an RF frequency f_{sweep}, tunable over 550 MHz. The phase of the mixing product was compared to that of a reference signal and the driving current of DFB-QCL2 was controlled to precisely tune the laser over 550 MHz while being phase-locked to one comb line of the referenced QCL-OFC.

3. Results and discussion

We measured the N_2O R(30) transition using a gas cell filled at 1.5 mbar. The comb referencing to a known transition and the knowledge of all RF frequencies determine the absolute frequency of DFB-QCL2 within the ambiguity given by the repetition rate of the comb. This ambiguity is lifted by the approximate knowledge of the transition of interest. The sweep over 550 MHz (point spacing of 0.55 MHz) took 20 s. We fitted the frequency-calibrated transmission curve using a Voigt profile which provides the transition center frequency [Fig. 2(b)].

We performed 10 measurements of the R(30) transition over the course of a week. Each measurement (of a duration of a few minutes) resulted in an absolute frequency value for the transition center with an uncertainty ranging from 126 kHz to 164 kHz (average 143 kHz). The main uncertainty source was the knowledge of the lock-point of DFB-QCL1 (on average 103 kHz). Our 10 measurements, shown in Fig. 2(c), agree with the literature values [1,2] within a 1-σ uncertainty and to each other within 2 σ, showing that the estimated uncertainties are coherent. The final uncertainty on the mean of our 10 values is 99 kHz, essentially limited by the accuracy of the N_2O R(8) transition frequency used as a reference (72 kHz) [1] and the cell pressure inaccuracy (60 kHz).

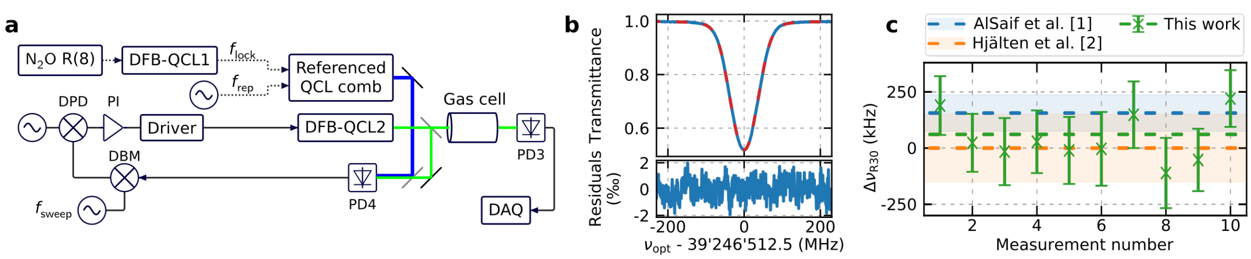

Fig. 2. Comb-calibrated spectroscopy. (a) Spectroscopic setup including the referencing to the comb (b) Transmission through the gas cell (blue) and Voigt fit (orange). Fit residuals in the lower panel. (c) Comparison of measured and literature values.

4. Outlook and conclusion

For the first time to the best of our knowledge, we have used a QCL-OFC referenced to an accurately-known molecular transition to perform comb-calibrated spectroscopy at 7.7 µm. We measured the frequency of the R(30) transition of N_2O to be 1309.122740(3) cm^{-1}, i.e., with a relative frequency accuracy of $3\cdot10^{-9}$, competing with state-of-the-art solutions based on nonlinear frequency conversion [1,2]. The main sources of uncertainty did not originate from the comb, such that the metrological limits of QCL-OFCs are still unknown. To conclude, we have demonstrated that QCL-OFCs can be used as highly-accurate frequency references for the MIR.

5. References

[1] 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 ν_1 fundamental band of $^{14}N_2{}^{16}O$," J. Quant. Spectrosc. Radiat. Transf. **211**, 172–178 (2018).

[2] A. Hjältén, M. Germann, K. Krzempek, A. Hudzikowski, A. Głuszek, D. Tomaszewska, G. Soboń, and A. Foltynowicz, "Optical frequency comb Fourier transform spectroscopy of $^{14}N_2{}^{16}O$ at 7.8 µm," J. Quant. Spectrosc. Radiat. Transf. **271**, 107734 (2021).

[3] J. Faist, G. Villares, G. Scalari, M. Rösch, C. Bonzon, A. Hugi, and M. Beck, "Quantum Cascade Laser Frequency Combs," Nanophotonics **5**, (2016).

[4] K. Komagata, A. Shehzad, M. Hamrouni, P. Jouy, F. Kapsalidis, M. Shahmohammadi, M. Beck, R. Matthey, V. J. Wittwer, J. Faist, T. Südmeyer, A. Hugi, and S. Schilt, "All-Mid-Infrared Stabilized Quantum Cascade Laser Frequency Comb with 30-kHz Frequency Stability at 7.7 µm," in *Conference on Lasers and Electro-Optics* (OSA, 2021), p. STu1H.3.