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High-Precision Mid-Infrared Spectroscopy with a Widely Tuneable SI-Traceable Frequency-Comb-Stabilised QCL

Dang Bao An Tran¹, Rosa Santagata¹, Mathieu Manceau¹, Anne Cournol¹, Louis Lecordier¹, Bérengère Argence¹, Oliver Lopez¹, Sean K. Tokunaga¹, Fabrice Wiotte¹, Haniffé Mouhamad¹, Andrei Goncharov¹, Michel Abgrall², Yann Le Coq², Hector Alvarez-Martinez², Rodolphe Le Targat², Won Kiu Lee², Dan Xu², Paul-Eric Pottie², Anne Amy-Klein¹, and Benoît Darquié¹

¹Laboratoire de Physique des Lasers, Université Paris 13, Sorbonne Paris Cité, CNRS, Villetaneuse, France

²LNE-SYRTE, Observatoire de Paris, Université PSL, CNRS, Sorbonne Université, Paris, France

Abstract— We report measurements of absolute frequencies of various polyatomic species around 10 μm , at level of accuracies ranging from 10 Hz to 10 kHz, using a widely tuneable SI-traceable optical frequency comb-stabilized quantum cascade laser.

I. INTRODUCTION

HERE is an increasing demand for precise molecular spectroscopy, in particular in the mid-infrared fingerprint window, whether it be for modelling our atmosphere, interpreting astrophysical spectra or testing fundamental physics. We present a high-resolution mid-infrared spectrometer traceable to primary frequency standards providing a unique combination of resolution, tuneability, detection sensitivity and frequency control.

II. EXPERIMENTAL SETUP

Fig. 1 shows the experimental setup, a mid-infrared spectrometer based on a widely tuneable, ultra-stable, and SI-traceable quantum cascade laser (QCL) working at 10.3 μm .

This set up comprises a tuneable and ultra-stable optical local oscillator (OLO) based on a 1.54 μm laser diode of frequency ν_{OLO} . Two sidebands are generated in the OLO signal using an electro-optic modulator (EOM) driven by a home-made phase-jump-free microwave synthesizer exhibiting a 9 GHz continuous tuning range. The upper sideband of frequency $\nu_{OLO} + f_{EOM}$ (with f_{EOM} the microwave frequency driving the EOM) is phase-locked to a 1.54 μm remote optical frequency reference signal of frequency ν_{ref} , using the phase-lock loop PLL1 shown in Fig. 1. This reference signal, generated at the French national metrological institute (LNE-SYRTE), is a 1.54 μm laser locked to an ultra-stable cavity and its absolute frequency is calibrated there against some of the world's best atomic clocks. It is then transferred to Laboratoire de Physique des Lasers through a 43-km long fibre link with active compensation of the propagation-induced phase noise. The OLO frequency is then given by

$$\nu_{OLO} = \nu_{ref} - f_{EOM} - \Delta_1, \quad (1)$$

with Δ_1 the frequency of the beat-note signal between $\nu_{OLO} + f_{EOM}$ and ν_{ref} .

An optical frequency comb (OFC) with two outputs centred at 1.55 and 1.85 μm is then used to transfer the spectral properties of the near-infrared reference signal to the QCL. We detect a beat-note signal between a comb tooth at frequency $p \times f_{rep} + f_{ceo}$ (with $f_{rep} \sim 250$ MHz the comb repetition rate, $p \sim 800\,000$ the comb mode integer, and f_{ceo} the comb offset frequency) and ν_{OLO} on photodiode PD2 (see Fig. 1). After

removing f_{ceo} , we obtain the resulting frequency beat-note signal at a frequency

$$\Delta_2 = \nu_{OLO} - p \times f_{rep}. \quad (2)$$

The comb repetition rate is stabilized by locking Δ_2 to a fixed radio-frequency signal (typically 150 MHz) using phase-lock loop PLL2.

Fig. 1. Experimental setup. OFC: optical frequency comb; QCL: quantum cascade laser; PLL: phase-lock loop; EOM: electro-optic modulator; SFG: sum frequency generation, PD: photodiode, DDS: direct digital synthesizer, DAQ: acquisition card.

Finally, the QCL frequency is phase-locked to a high-order harmonic of the comb repetition rate. To this end, we perform sum-frequency generation between the comb output beam at 1.85 μm (comb mode frequency $q \times f_{rep} + f_{ceo}$ with $q \sim 680\,000$) and the QCL beam, which results a shifted comb centred at 1.55 μm with comb mode frequencies $q \times f_{rep} + f_{ceo} + \nu_{QCL}$. In order to stabilize the QCL frequency, a beat-note signal at frequency $\Delta_3 = (p - q) f_{rep} - \nu_{QCL}$ between the shifted comb and the original comb is detected on photodiode PD3. The QCL frequency is finally stabilized by locking this beat-note signal to a fixed radio frequency signal (typically 11 MHz) using phase-lock loop PLL3. The QCL frequency is given by the following equation,

$$\nu_{QCL} = n \times f_{rep} - \Delta_3 = \frac{n}{p} (\nu_{ref} - f_{EOM} - \Delta_1 - \Delta_2) - \Delta_3, \quad (3)$$

where $n = p - q \sim 120\,000$.

The experimental setup therefore allows the stability and SI-

traceability of the optical frequency reference, and the tuneability of the OLO to be transferred to the QCL. This results in a ~ 0.1 Hz QCL linewidth, a stability below 10^{-15} from 0.1 to 10 s and an uncertainty on the absolute frequency below 4×10^{-14} [1,2]. The spectral range covered by our spectrometer is ~ 100 GHz limited by the spectral window of the QCL used. Scanning the OLO carrier frequency over 9 GHz allows the QCL frequency to be tuned over more than 1 GHz.

III. RESULTS

As illustrated in Fig. 1, we have used the apparatus to carry out ultra-high resolution spectroscopy of polyatomic molecules in a compact multipass absorption cell and few-meter-long Fabry-Perot cavities. We use frequency modulation to improve detection sensitivity.

In order to demonstrate the apparatus' potential for state-of-the-art high-precision spectroscopic measurements, we have conducted saturated absorption spectroscopy of OsO_4 using a Fabry-Perot cavity [1]. Fig. 2 shows a sub-Doppler spectrum of two rovibrational transitions of OsO_4 averaged over ten up and down scans. It is recorded using frequency modulation and third-harmonic detection. The QCL frequency is swept by scanning the frequency of the synthesizer used to phase-lock the QCL to the OFC. Typical experimental parameters are a pressure of 0.04 Pa, an intra-cavity power of 50 μW , and a beam diameter of 6 mm. The spectrum exhibits a peak-to-peak linewidth of ~ 25 kHz and a signal to noise ratio of 200. The data is fitted to a combination of a third and a fifth derivative of a Lorentzian to account for the line shape deformation caused by the choice of frequency modulation parameters. The line-centre frequencies of these transitions and others were determined at a level of accuracy ranging from 20 to 40 Hz (see [1] for more details).

Fig. 2. Saturated absorption spectrum of OsO_4 recorded using a Fabry-Perot cavity. Two lines are recorded over a span of 5 MHz in steps of 5 kHz, using frequency modulation and third harmonic detection. Inset: spectrum of the reference line recorded over 200 kHz in steps of 1 kHz. Taken from [1].

As illustrated in Fig. 3, we have also carried out saturated absorption spectroscopy of several rovibrational transitions of methanol using a multipass absorption cell. Typical experimental conditions are a pressure of 1 Pa, an average intracell power of ~ 1 mW, a laser beam waist of ~ 1 mm and first-harmonic detection. The QCL frequency is swept by scanning the EOM frequency and in turn the OLO carrier frequency. Fig. 3(b) displays a sub-Doppler spectrum of the $P(E,co,0,2,33)$

rovibrational line of methanol averaged over only one up and down scan. It exhibits a peak-to-peak linewidth of ~ 400 kHz and a signal-to-noise ratio of 310. The solid yellow line is a fit to the data (see [2] for details of the line shape used). The line-centre frequency of the transition was determined with a statistical uncertainty of ~ 1 kHz and sub-10 kHz global uncertainty. Fig. 3(b) demonstrates the unprecedented continuous tuning capability resulting from using our widely tuneable and ultra-stable OLO. It shows a sub-Doppler spectrum of methanol around 971.565 cm^{-1} spanning 400 MHz and exhibiting five rovibrational transitions of methanol [2].

Fig. 3. (a) Saturated absorption spectrum of the $P(E,co,0,2,33)$ rovibrational transition of methanol, with fit and residuals. (b) Saturated absorption spectrum of methanol spanning 400 MHz. Taken from [2].

To conclude, we have developed an apparatus showing an unprecedented combination of wide tuneability, high sensitivity, and spectral resolution and used it to carry out saturated absorption spectroscopy. We have demonstrated record uncertainties ranging from ~ 10 Hz to 10 kHz on rovibrational absolute frequencies of various species of interest for space and atmospheric physics, and for testing fundamental physics, including osmium tetroxide, methanol, ammonia, trioxane... This confirms our apparatus' potential for our ongoing precise spectroscopic measurements with molecules [3].

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