Comb-assisted spectroscopy of CO₂ absorption profiles in the near- and mid-infrared regions

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Abstract A method of performing comb-assisted spectroscopy in the 2- and 4- μ m wavelength regions is deeply discussed and applied to the spectroscopic characterization of absorption lines of a CO₂ gas sample. The method relies on the use of a sum-frequency-generation scheme to relate the frequency of the probing laser to a frequency comb provided by an Er:fiber femtosecond oscillator. Controlled scans of the comb repetition frequency allow highly accurate and reproducible acquisition of the absorption spectra, which are used for subsequent retrieval of the underlying spectroscopic parameters.

1 Introduction

Climate modeling and global change research programs are setting unprecedented accuracy targets in gas-sensing missions for atmospheric CO₂ and other greenhouse gases. For instance, the Orbiting Carbon Observatory (OCO), which is the satellite sensor expressly developed by NASA for atmospheric measurements of CO₂ concentration, was born with

the ambitious goal of providing data with a precision better than 0.5 % [1–3]. High-quality spectroscopic parameters, traditionally used to describe absorption spectra, represent an indispensable prerequisite for the achievement of the desired accuracy levels. In turn, the accurate determination of positions, strengths, and pressure-broadened widths of the individual spectral lines requires a detailed and thorough understanding of the near- and mid-infrared spectral line shapes of CO₂ and other radiatively important trace gases.

In the last few years there has been a growing interest toward the development of highly accurate spectroscopic methods for the measurement of molecular line profiles [4]. An interesting solution has been recently implemented by Castrillo et al. using the technique of offset-frequency locking of a pair of extended cavity diode lasers in the near infrared [5]. In such a system, the master laser is frequency stabilized against a narrow sub-Doppler molecular line, while the slave laser is actively controlled so that its emission frequency maintains a given and controlled offset with respect to the frequency of the master laser. This method allows the performance of highly accurate and reproducible frequency scans of the slave laser around a given vibration–rotation line, thus ensuring an extremely high spectral fidelity, limited only by the noise level [6]. It exhibits, however, two main drawbacks: (i) it can be applied only to those absorption lines that are a few GHz away from a sufficiently strong line to be used for the absolute frequency stabilization of the master laser; (ii) its extension to the mid infrared is surely not straightforward, as a fast detector with a bandwidth larger than 1 GHz is required, in order to monitor the beat note between the two lasers.

Invented nearly 11 years ago, optical frequency combs have led to breakthrough advances in the field of frequency metrology [7, 8], enabling a direct and phase-coherent link

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of visible and near-infrared frequencies to the microwave portion of the electromagnetic spectrum and, hence, to the primary frequency standard. Based upon mode-locked femtosecond lasers, optical frequency combs provide hundreds of thousands of sharp, equally spaced, spectral components, distributed across hundreds of nanometers, and each tooth can act as an absolute frequency reference. As a consequence, it is clear how an optical frequency comb could play the role of thousands of master lasers in spectroscopic schemes similar to that described above. In the framework of molecular spectroscopy, frequency combs have been so far mostly applied to the determination of line-center frequencies with extreme accuracy [9–13]: in such measurements the frequency comb typically acts as a rigid ruler to count the frequency of a continuous-wave (cw) laser that has been tightly locked to a sub-Doppler absorption feature. Less attention has been paid to measurement schemes devoted to the acquisition of the full absorption profile, which is indeed essential both for the quantitative determination of the gaseous samples and for a full understanding of the collisional processes affecting the line shape. In such context, acetylene only has been to date thoroughly investigated in the near infrared (NIR) [14–17], while in the mid infrared (MIR) the examples encompass a few measurements on CH₄ [18, 19] at 3 μm and one measurement on CO₂ at 4.4 μm [20], the latter one being however performed in a free-running regime without phase lock between the comb and the probing quantum-cascade laser.

In this paper we describe a pair of examples of frequency-comb-assisted spectroscopy of CO₂, in the near- and in the mid-infrared spectral regions, where the entire absorption profile is acquired with high accuracy. The investigated lines are the P15f line of the $2\nu_2^2 + \nu_3 - 2\nu_2^2$ band at 4.3 μm and the P28f line of the $2\nu_1 + \nu_2^1 + \nu_3 - \nu_2^1$ band at 2.09 μm . To probe these lines a quantum cascade laser (QCL) and a solid-state (Tm:Ho:YAG) laser at 2.09 μm were respectively used in combination with an Er:fiber-based frequency comb with variable repetition rate. The frequencies of both probing lasers have been referred to integer multiples of the comb repetition frequency, without contribution from the carrier-envelope offset frequency, by means of a suitable sum-frequency-generation scheme in a PPLN crystal.

2 Sum-frequency generation for absolute referencing of mid- and near-infrared frequencies

As pioneered in some early papers on MIR comb-assisted spectroscopy [21–23], mid-infrared frequencies emitted by single-mode narrow-line-width lasers can be referred to NIR frequency combs through nonlinear optical processes such as difference-frequency generation (DFG) or sum-frequency generation (SFG). In the DFG case the difference

between two spectral portions of an octave-spanning NIR comb produces an offset-free MIR comb spectrally overlapped to the cw probing laser, whose frequency can then be determined by measuring the beating signal with the nearest comb mode. In the SFG case, hereafter described in more detail, the frequency ν_{cw} of the laser is added to a given spectral portion of a NIR comb described by the relation $\nu_1 = f_{\text{ceo}} + n \cdot f_{\text{rep}}$ to generate a frequency-shifted comb described by $\nu_{\text{sf}} = f_{\text{ceo}} + n \cdot f_{\text{rep}} + \nu_{\text{cw}}$ (f_{ceo} and f_{rep} being the carrier-envelope-offset frequency of the comb and the laser repetition frequency, respectively). The superposition of ν_{sf} with a high-frequency spectral portion $\nu_2 = f_{\text{ceo}} + m \cdot f_{\text{rep}}$ of the original comb produces a beat note at frequency $f_{\text{beat}} = |\nu_{\text{sf}} - \nu_2|$ that can be used to refer ν_{cw} to the comb repetition rate by the relationship $\nu_{\text{cw}} = \pm f_{\text{beat}} + (m - n) \cdot f_{\text{rep}}$, without any contribution from the comb offset frequency f_{ceo} . The advantage of the SFG approach is that the beating occurs in the NIR, where the bandwidth and noise level of the detectors are not traded off by high costs or by the need of cryogenic cooling.

Figure 1 reports the spectra of the frequency combs that have been experimentally used to measure the absolute frequencies of cw optical sources operating at around 4.3 μm (a) and at 2.09 μm (b), respectively, a quantum cascade laser (QCL) and a Tm:Ho:YAG laser, starting from the two amplified and phase-coherent outputs of a femtosecond Er:fiber oscillator. The green line refers to the spectrum of the octave-spanning supercontinuum (SC) generated from a highly nonlinear fiber seeded by one of the laser outputs. By tuning the chirp of the pulses injected in the fiber, the SC spectral extent can be manipulated and stretched to attain a maximum of ~ 150 -THz bandwidth, roughly corresponding to the 1030–2100 nm range. As such, any cw frequency lower than 150 THz—or wavelength longer than 2100 nm—can in principle be referred to the comb by means of a suitable SFG process. The efficiency of such process remains however rather poor if the ν_1 comb—the one that is added to ν_{cw} —is directly extracted from the SC without any additional amplification. An amplified ν_1 comb can be obtained by exploiting either the second amplified laser output at 1.55 μm (a) or the external amplification of the 2.05- μm spectral portion of the SC (b). Optical frequencies as high as 100 THz (the frequency difference between 1.55 μm and 1.03 μm) and 150 THz can be measured in cases (a) and (b), respectively. The main drawback of the second configuration is the need of an external amplifier; in our experimental setup we employed a home-built solid-state Ho:YLF amplifier in a multipass configuration [24].

Figure 1 also reports the experimental spectra produced by the SFG processes, corresponding to the ν_{sf} combs. In such processes, a single-mode cw laser (at 4.3 and 2.09 μm for cases (a) and (b), respectively) and an amplified pulse train at 100 MHz (centered at 1.55 and 2.05 μm , respectively) are collinearly recombined in a PPLN crystal with an

Figure 1 consists of two plots, (a) and (b), showing Intensity versus Wavelength (nm). Plot (a) shows the full spectrum from 1100 to 4500 nm. The legend indicates four components: Amplified Comb (red), SF Comb (blue), QCL (black), and SC (green). The spectrum shows a broad peak around 1550 nm labeled v_n and a sharp peak at 4250 nm labeled v_{CW} . Plot (b) is a zoomed-in view of the 1000-2200 nm range. The legend includes Amplified Comb (red), SF Comb (blue), Tm-Ho (black), and SC (green). The spectrum shows a broad peak around 1050 nm labeled v_m , a sharp peak at 1000 nm labeled $v_{SF,n}$, and a sharp peak at 2050 nm labeled v_n . The v_{CW} peak is also visible at 4250 nm.

[illegible]
$$\Delta\left(k_{\text{sf}} - k_1 - k_{\text{cw}} - \frac{2\pi}{\Lambda}\right) = \left[\left(\frac{\partial k}{\partial \omega}\right)_{\text{sf}} - \left(\frac{\partial k}{\partial \omega}\right)_1\right]\Delta\omega = [\tau_{\text{g, sf}} - \tau_{\text{g, 1}}]\Delta\omega, \quad (1)$$

For the estimation of the optical power carried by the sum-frequency beam, we multiplied the number N of comb modes encompassed by the experimental $\Delta\nu_{\text{FWHM}}$, i.e. 31500 and 710 for cases (a) and (b), respectively, by the power expected from the interaction between the cw laser and an individual mode of the comb (see e.g. Ref. [25] for SFG conversion efficiency). With optical powers of 770 μW and 10 mW for the cw lasers, of 1.5 μW and 35 μW for the individual comb modes, and a spot size of 25 μm , the average power carried by the sum-frequency beams can be calculated to be 13.5 nW and 710 nW for cases (a) and (b), respectively, in reasonable agreement with the measured 5 nW and 150 nW. The discrepancy is mainly due to the plane-wave approximation adopted for the calculation of the sum-frequency power.

3 Experimental setup

Figure 2 shows the experimental setup for comb assisted spectroscopy in the 2 μm and 4 μm wavelength regions. The Er:fiber femtosecond oscillator is a dual-branch version of the Toptica FFS model including piezoelectric stages and locking electronics to achieve a continuous scan of the repetition rate over the 99.9–100.1 MHz range. One output delivers nearly 250 mW at 1.55 μm , while the second output provides nearly 160 mW of SC, whose spectral extension can be tuned by modification of the chirp of the pulses injected in the nonlinear fiber. The short-wavelength part of the SC, which acts as the comb at frequency ν_2 , is extracted by a dichroic mirror and then superimposed to the comb at the sum frequency ν_{sf} after suitable delay. The long-wavelength part of the SC is sent to a Ho:YLF multipass amplifier providing amplification of the 2.05- μm spectral portion up to a power spectral density of $\sim 100 \mu\text{W}$ per mode of the comb. Such an amplified 2.05- μm pulse train, or to the user's choice the 1.55- μm laser output (power spectral density of $\sim 2.5 \mu\text{W}$ per comb mode), acts as the comb at frequency ν_1 and is used to seed the SFG process with the cw laser. The nonlinear interaction between the two lasers occurs in a PPLN crystal slightly tilted to avoid backreflection-induced instabilities of the cw laser beam. Once collimated, the light exiting the crystal is heterodyned with the ν_2 comb after proper spectral filtering, which is indeed necessary to reject the light not involved in the beating process. The beat-note signal used for locking the laser to the comb is collected by an amplified InGaAs detector (model 1811-FS, New Focus) with a 125-MHz bandwidth, a NEP of $2.5 \text{ pW/Hz}^{0.5}$, and a saturation power of $55 \mu\text{W}$.

The spectroscopic measurements are performed by sending a small fraction of the cw laser beam to a cell containing CO_2 at variable pressures, and by acquiring the transmitted optical power against the laser frequency. For the absorption measurements at 4.3 μm , we used a cw liquid-nitrogen-cooled distributed feedback (DFB) laser from Alpes Lasers providing single-longitudinal-mode operation and an optical power up to 20 mW, with a threshold of 90 mA and a slope of $170 \mu\text{W/mA}$. It was driven by a commercial power supply (model LDC-3744B, ILX Lightwave) with a rather high noise figure of 10 μA rms. The CO_2 cell was 12-cm long and the output power was monitored with a Peltier-cooled MCT photovoltaic detector (250- μm diameter, nearly 2-MHz bandwidth, Judson Technologies). For the measurements at 2.09 μm , a single-frequency Tm-Ho:YAG laser with mode-hop-free tunability of about 1.2 GHz, coarse tunability of about 10 nm, and a maximum output power of 20 mW was used. Due to the fact that CO_2 is a quite heavy molecule, a strong scaling of the line strength occurs when moving from the fundamental absorption band at 4.3 μm to the overtone band at 2.09 μm : for this reason a multipass

Fig. 3 Electrical spectrum of the beat note between comb and QCL (blue line) and comb and Tm-Ho:YAG laser (red line) in phase-locked conditions, with a resolution bandwidth of 300 kHz (QCL) and 10 kHz (Tm-Ho:YAG laser)

Herriott cell with an equivalent absorption path of 33 m was adopted in this spectral region.

Comb referencing of the probing laser is implemented by phase locking the beat note to a radio-frequency signal generated from a synthesizer referenced to a GPS-disciplined rubidium microwave standard (model 4410A, Symmetri-com). The phase lock is achieved by combining a 200-kHz-bandwidth phase detector (model PDF-200W, RFBay) with a FPGA-based PID servo acting either on the QCL pump current or on the end-cavity piezo mirror in the case of the Tm-Ho:YAG laser. Spectral analysis of the error signal at the output of the phase detector revealed in both cases a control bandwidth at the kHz level, as limited by the QCL current driver in one case and by the piezo transducer in the other. Absolute scanning of the cw laser frequency is achieved by changing the comb repetition rate around 100 MHz with a radio-frequency synthesizer also synchronized to the GPS-disciplined rubidium oscillator. Thanks to the much broader mode-hop-free tuning interval, the QCL could be robustly scanned over repetition rate tuning ranges as high as 30 kHz, corresponding to more than 20 GHz in the optical domain (55 mA of current modulation). We experimentally verified that without phase lock to the comb, direct modulation of the QCL current resulted in a highly nonlinear and not reproducible frequency axis, mostly due to temperature variations of the QCL chip. The overall measurement chain is fully controlled by a PC and synchronized with the repetition rate for the acquisition of absorption profiles with absolute frequency calibration.

4 Results and discussion

Figure 3 shows the beating signals between comb and cw laser in the two cases considered. The QCL was found to exhibit a rather broad line width of ~ 7 MHz, primarily due to the current noise contribution [26]. For this reason, the offset frequency chosen for the comb-to-the-QCL phase lock

Fig. 4 *Left-hand panels:* CO₂ absorption profiles as a function of absolute frequency at 4.3 μm (a) and 2.09 μm (b), as averaged over five scans lasting 2 min each. *Right-hand panels:* line-center frequencies retrieved from the fitting of several independent acquisitions, as reported with respect to a relative frequency scale

was kept at a prudential value of 15 MHz. A lower 10-MHz value was adopted for the solid-state Tm-Ho:YAG laser, which was found to exhibit a particularly narrow line width of about 76 kHz, comparable to the expected line width of the individual comb modes [27]. Comb-assisted frequency scans of the two lasers allowed absorption profiles to be acquired with excellent reproducibility of the frequency axis and with signal-to-noise-ratios as high as 1500 at 4.3 μm and 1800 at 2.09 μm , as evaluated from the rms deviation between the experimental points and a fitting curve calculated according to a Galatry profile, as adopted to take into account the experimental evidence of a line-narrowing effect. These values refer to 10-min-long acquisition times, as composed by five repeated scans lasting 2 min each. Figure 4 shows the measured absorption profile for two selected lines of the main CO₂ isotopologue: the P15f rovibrational line of the $(0, 2^2, 0) \rightarrow (0, 2^2, 1)$ band at $\sim 4.329 \mu\text{m}$ (panel a), at a pressure of 7.3 Torr (as measured with a relative uncertainty of 2.5×10^{-3}) and at a temperature of 296 K (accuracy of 0.1 K), and the P28f line of the $(0, 1^1, 0) \rightarrow (2, 1^1, 1)$ band at 2.092 μm (panel b), at 10 Torr and 298 K.

The least-square fitting procedure applied to the experimental molecular spectra provides the repetition rate value $\langle f_{\text{rep}} \rangle$ in correspondence with the line center. To move to the absolute optical frequency, the correct value of the $(m - n)$ repetition rate multiple as well as the correct sign of f_{beat} have to be assigned in the formula $\nu_{\text{cw}} = \pm f_{\text{beat}} + (m - n) \cdot \langle f_{\text{rep}} \rangle$. This was achieved by employing a wavemeter (instrumental accuracy of 40 MHz) for the P28f line, and exploiting the nominal line center frequency value reported in HITRAN for the P15f line (30 MHz of accuracy). Such a calibration is sufficient for an unambiguous evaluation of the $(m - n)$ term, while the correct sign of f_{beat} was assessed in a free-running regime by monitoring the sign of the shift of the beating note upon changing the repetition rate.

In the left-hand panels of the two figures, with an enlarged frequency scale, the line-center values retrieved af-

ter repeated independent measurements are reported for the two lines. Even if the two measurements exhibit a similar signal-to-noise ratio, the rms deviation is 227 kHz for case (a) and 1 MHz for case (b). Such difference is mainly due to the fact that at 2 μm the line contrast is nearly two times lower, and the higher $(m - n)$ factor (~ 2) amplifies the effect of the frequency noise of the electrical waveform synthesizer. To a lesser extent the larger rms deviation at 2 μm is related to the ~ 1.5 times larger absorption line width and to the limited mode-hop-free tunability of the Ho:YAG laser, with the difficulty of covering the wings of the absorption profile. When averaging over the different line center frequency determinations, an absolute value of $2311.756144(8) \text{ cm}^{-1}$ ($69.30470567(3) \text{ THz}$) is found for the P15f line and of $4784.398630(7) \text{ cm}^{-1}$ ($143.4326625(2) \text{ THz}$) for the P28f line, differing by less than 1.7 and 0.2 MHz from the respective HITRAN values: $2311.756200 \text{ cm}^{-1}$ and $4784.398625 \text{ cm}^{-1}$. A remarkable precision below 10^{-9} is thus achieved with a reasonably simple experimental method. By repeating spectra acquisition at variable pressures and with calibrated cell lengths, the method proves useful to retrieve other spectroscopic parameters such as line intensity factors and pressure-shift and pressure-broadening coefficients, as recently reported in Ref. [31].

5 Conclusions

This paper shows how to exploit a sum-frequency-generation process to achieve phase lock between an Er: fiber-based frequency comb and cw lasers operating in the infrared at wavelengths above 2.05 μm . Such an apparatus holds a great potential to extend comb-assisted spectroscopy to the MIR spectral range, where few realizations have been reported so far. The proposed configuration, here applied at 4.3 and 2.09 μm , is likely to be scalable to higher wavelengths provided a sufficiently high conversion efficiency is achieved

in the SFG process. To this purpose it is worth recalling that Refs. [28] and [29] show that, starting from the same laser source used here, wavelengths between 3.2 and 4.8 μm can be easily achieved by DFG in PPLN with power levels in the 300 μW to 1 mW range, and wavelengths from 5 to 12 μm are accessible by DFG in GaSe crystals in the 100–200 μW range using comparable incoming powers. In both cases the 1.55- μm laser output was made to interact with selected spectral portions of the SC output, according to an approach that is dual with respect to the SFG approach here reported. Those data, as combined with the fact that commercially available QCLs now exist with narrower line width [30] and optical power in excess of the sub-10-mW level exploited here, make wavelength scalability a feasible achievement in the whole fingerprint region.

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