Mid-infrared dual-comb spectroscopy with GHz resolution using soliton microcombs

Chengying Bao,^{1,†} Zhiquan Yuan,^{1,†} Lue Wu¹, Myoung-Gyun Suh¹, Qiang Lin², and Kerry Vahala^{1,*}

¹ T. J. Watson Laboratory of Applied Physics, California Institute of Technology, Pasadena, CA 91125, USA.

² Department of Electrical and Computer Engineering, University of Rochester, Rochester, NY 14627, USA

[†]These authors contributed equally to this work.

*vahala@caltech.edu

Abstract: Microcomb based dual-comb spectroscopy of methane in the mid-infrared is demonstrated with GHz resolution. This fine resolution is enabled by generating spectrally densified mid-infrared combs via interleaved difference-frequency-generation. © 2021 The Author(s)

Dual-comb spectroscopy (DCS) is a powerful technique for precision measurement of gas spectra [1]. And soliton microcombs have recently been used to implement this method [2]. Ultimately, chip-based DCS modules using microcombs could revolutionize field measurement of various gases including chemical threats and greenhouse gases such as CO₂ and CH₄. In the application of DCS to gases, it is favorable to operate in the mid-infrared (mid-IR) where the gases have much stronger absorption [3]. And mid-IR silicon microcomb DCS has been reported with a line spacing of 127 GHz [4]. Such a large line spacing is a result of the small microresonator form factor and causes spectral under-sampling of gas absorption lines whose bandwidths are usually narrower than tens GHz. Attaining direct generation of narrow line spacing microcombs in the mid-IR has so far not been possible. Interleaved difference-frequency-generation (iDFG) is a recently reported alternative approach for generation of mid IR comb spectra [5]. By selecting two near-infrared (near-IR) combs having repetition rates f_r and $(N-1)f_r/N$, a temporal interleaving effect upon difference frequency generation is created, and the resulting mid-IR comb is densified to a line spacing of f_r/N .

Here, we use iDFG densified microcombs for mid-IR DCS with GHz resolution. A pair of counter-propagating (CP) solitons with repetition rates of f_r^{cw} and f_r^{ccw} were generated in a 22 GHz silica wedge microresonator [6]. The locking dynamics between CP solitons that stabilize $\Delta f_r = f_r^{\text{cw}} - f_r^{\text{ccw}}$ [6] enhance the coherence of the resulting DCS system. The two soliton streams were then photodetected to drive a pair of electro-optical combs (EO-combs) at the rates of $(N-1)f_r^{\text{cw}}/N$ and $(N-1)f_r^{\text{ccw}}/N$ ($N = 8$ or 16) for iDFG (see experimental setup in Fig. 1(a)). The two near-IR combs operated at $1.55 \mu\text{m}$ (soliton microcomb) and $1.06 \mu\text{m}$ (EO-comb), thus generating mid-IR combs in the $3.3 \mu\text{m}$ upon iDFG.

Fig. 1. (a) Experimental layout for iDFG DCS. PM (IM): phase (intensity) modulator, AOM: acousto-optical modulator, WDM: wavelength division multiplexer, circ.: circulator. (b) The mid-IR interferogram of conventional DFG and iDFG ($N = 8$). (c) The corresponding RF spectra. (d) Zoom in of the spectra in panel c for a RF bandwidth of about Δf_r or 22 GHz optical bandwidth.

Fig. 2. (a) DCS of methane Q branch using conventional DFG and iDFG $N = 8$. Hitran spectra are also provided. (b) DCS of methane P(3) and ethane using iDFG with $N = 8$ and $N = 16$. The right panels of (a, b) are zoom-in of the spectra. (c) Measurement precision of the methane concentration for iDFG $N = 8$. The inset shows the sequence of methane concentration measured within 0.5 ms for P(3) branch using iDFG $N = 8$.

The 1.06 μm EO-comb was seeded by a tunable external cavity diode laser and its center wavelength was fine adjusted to probe different methane absorption branches. The soliton microcomb and the EO-comb were combined to pump a PPLN crystal for iDFG. The generated mid-IR combs were combined in free space to pass through a 5 cm long gas cell with a mixture of 2% methane and 0.5% ethane (buffered by nitrogen to a pressure of 760 torr). Their detected interferogram (without cell) was recorded by an oscilloscope. The interferograms for conventional DFG (continuous wave input at 1.06 μm with EO-combs turned off) and iDFG are shown in Fig. 1(b). The interferogram repeats at the rate of Δf_r (about 80 kHz in experiment) for conventional DFG and at a rate of $\Delta f_r/N$ when iDFG is used. Transforming the interferograms into the frequency domain yields the RF spectra shown in Fig. 1(c). Spectral zoom-in illustrates the densification of the line spacing (Fig. 1(d)). The measured RF spectra also exhibit high SNR for iDFG with the peak SNR reaching $6800/\sqrt{5}$.

The gas absorption spectra were measured by normalizing the dual-comb spectra obtained with the gas cell by the spectra obtained without the gas cell. The measurement time is 200 ms. We first measured the Q branch of methane around 3015 cm^{-1} . The DCS results using DFG and iDFG ($N=8$) are shown in Fig. 2(a). The iDFG case enables measurement of the fine structure within the Q branch with a resolution of about 2.8 GHz, while the conventional DFG case strongly undersamples the absorption features. Similarly, the mixed absorption spectra of methane and ethane around 2980 cm^{-1} were also measured using iDFG DCS (Fig. 2(b)). The system can be reconfigured for different resolutions by selecting different N in the iDFG process. For example, using $N=16$ to produce a 1.4 GHz mid-IR comb enables better resolution of the ethane absorption, which has a narrower linewidth than methane (see zoom-in of Fig. 2(b)). The measurement precision was evaluated by analyzing the Allan deviation of the measured methane concentration in the 5 cm cell (concentration measured in 0.5 ms used as a single data point, see inset of Fig. 2(c)). The analysis shows that a measurement precision of 2.8 ppm·m was reached in 64 ms averaging time.

In summary, we demonstrated mid-IR DCS of methane with GHz resolution using the iDFG method. CP soliton microcombs in combination with EO-combs were employed. The system still uses a considerable amount of fiber optic components. However, a fully integrated device is potentially feasible. For example, the soliton microcomb, EO-modulators and PPLN waveguides have all been demonstrated using thin-film LiNbO₃ devices.

This work was supported by the DTRA (HD-TRA11810047), the AFSOR (FA9550-18-1-0353), the Resnick Institute at Caltech, and the Kavli Nanoscience Institute.

References

1. I. Coddington, N. Newbury and W. Swann, *Optica* **3**, 414 (2016).
2. M-G. Suh, Q-F Yang, K. Y. Yang, X. Yi, and K. Vahala, *Science*, **354**, 600-603 (2016).
3. G. Ycas, et al., *Nat. Photon.* **12**, 202 (2018).
4. M. Yu, Y. Okawachi, A. Griffith, N. Picqué, M. Lipson, and A. Gaeta, *Nat. Commun.*, **9**, 1869 (2018).
5. C. Bao, Z. Yuan, H. Wang, L. Wu, B. Shen, K. Sung, S. Leifer and K. Vahala, *Optica*, **6**, 309 (2020).
6. Q. F. Yang, X. Yi, K. Y. Yang, and K. Vahala, *Nat. Photon.*, **11**, 560 (2017).