Nanophotonic supercontinuum based mid-infrared dual-comb spectroscopy

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Abstract: We demonstrate a broadband mid-infrared dual-comb spectroscopy for parallel gas-phase detection in the functional group region from $2800-3600$ cm^{-1}, using dispersion engineered silicon nitride dual-core waveguides which produce broadband, intensity-enhanced and coherent mid-infrared frequency combs.

OCIS codes: (320.6629) Supercontinuum generation, (300.6340) Spectroscopy, infrared

Introduction — Mid-infrared (mid-IR) frequency combs are of great interest for molecular spectroscopy. They not only provide a broadband and coherent laser source to characterize molecular vibrations as strong absorption in the mid-IR spectral range ($2.5-10$ μm), but also can constitute superior spectroscopic applications, in particular the dual-comb spectroscopy that allows for rapid and high-resolution detection [1].

In addition to conventional approaches such as mid-IR mode-locked lasers and different frequency generation (DFG) [2], recent work has also highlighted mid-IR frequency comb generation from compact and chip scale platforms. In particular, photonic chip-based supercontinuum process can lead to efficient mid-IR frequency combs [3]. This approach can be seeded in the telecom band (i.e. 1.55 μm) as well as at 2 μm [4], which is the most accessible range for femto-second fiber laser technologies. By means of dispersion engineering that is enabled by the lithographically controlled waveguide geometry, mid-IR dispersive wave can be precisely tailored and enhanced in terms of both the intensity and the bandwidth, performing an efficient frequency comb that inherits the full coherence of the seeding laser. With a simple configuration, photonic supercontinuum based mid-IR frequency comb can be readily extended for dual-comb spectroscopy.

Here, we demonstrate a broadband mid-IR dual-comb spectroscopy for parallel gas phase detection in the $2.5-3.7$ μm spectral range, using photonics chip based supercontinuum in Si_3N_4 waveguides seeded in the telecom band. We demonstrate a novel scheme of dispersion engineering that allows for broadband mid-IR generation, and we showcase the parallel gas-phase detection of both methane (CH_4) and acetylene (C_2H_2).

Results — The experimental setup is shown in Fig. 1(a). The seeding source is a pair of fully stabilized fiber laser based frequency combs (*Menlo Systems*) centered at the wavelength of 1.55 μm. The comb mode spacing (i.e. the repetition rate) is ~ 250 MHz. Both combs are mutually locked, with one comb tooth in each optically locked to a continuous wave laser as the reference. This allows for a tiny difference in the comb repetition rate, i.e. ~ 320 Hz, such that a large optical range is effectively covered by the dual-comb spectrometer. Both combs are amplified such that they produce femto-second and high-intensity laser pulses (duration < 70 fs, maximal averaged power ~ 400 mW) to seed the mid-IR supercontinuum in two chip-based Si_3N_4 waveguides.

The Si_3N_4 waveguides are fabricated by means of the photonic Damascene-reflow process [5]. In contrast to a conventional single-core waveguide, here we use a dual-core waveguide structure that provides more degrees of freedom in controlling the waveguide geometry, and allows for advanced dispersion engineering. In particular, mode hybridization will occur in such waveguides, which leads to a pair of symmetric and asymmetric modes and locally alters the waveguide dispersion. In principle, this effect can be at arbitrary wavelength range. When operating in the mid-IR, it can lead to more efficient and broadband mid-IR generation compared with single-core waveguides [6]. As a result, indeed a broadband mid-IR frequency comb generation covering the $2.0-3.7$ μm spectral range is accomplished, see Fig. 1(b). The output mid-IR power is typically > 1 mW (cut-on at 2.5 μm) from each waveguide.

At the output side, one mid-IR beam serves as the detecting arm and passes through a gas cell (length 104 cm) which is filled with methane (CH_4) at 430 ppm and acetylene (C_2H_2) at 420 ppm and nitrogen (N_2) as buffer gas, with total pressure 1 atm. With both spatial and spectral overlap between the detecting and the reference arms, the

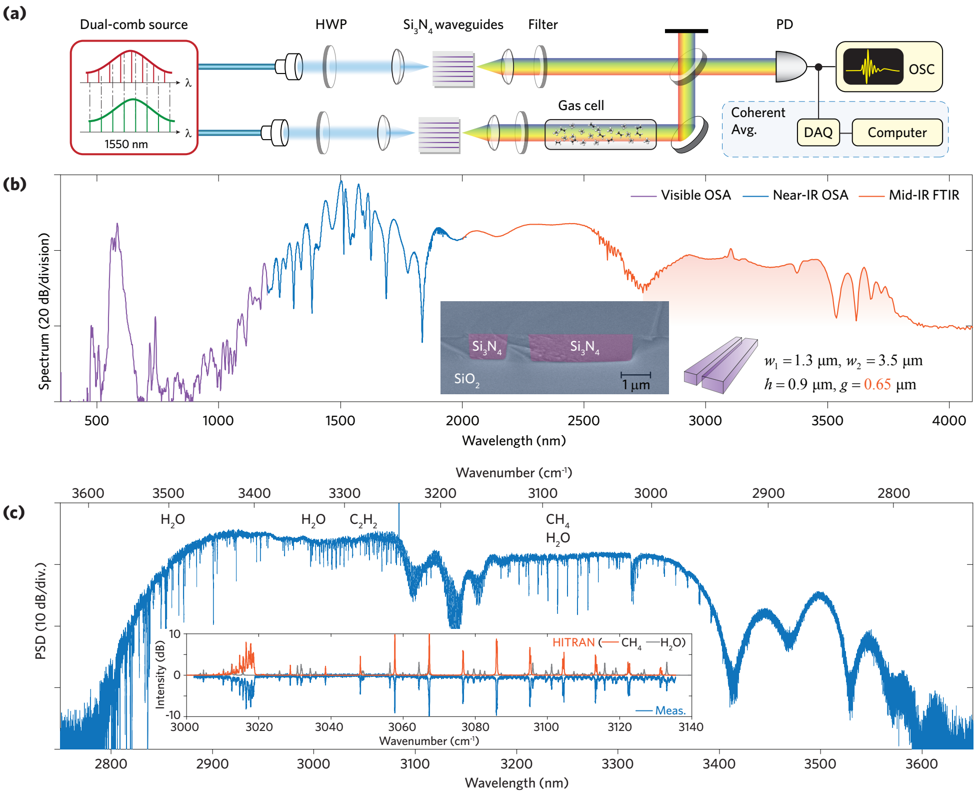

Fig. 1. (a) Schematic setup for photonic supercontinuum based mid-IR dual-comb sepctroscopy. HWP: half-wave plate; PD: photo-detector; OSC: oscilloscope; DAQ: data acquisition unit. (b) Full spectrum of the supercontinuum in the dual-core Si_3N_4 waveguide, where the width of the dual cores is $w_1 = 1.3\ \mu$m and $w_2 = 3.5\ \mu$m, respectively. The gap distance in between the cores is $g = 0.65\ \mu$m. The waveguide height is $h = 0.9\ \mu$m. Inset: false colored SEM picture of the cross section of the dual-core Si_3N_4 waveguide. (c) Retrieved mid-IR frequency comb featuring absorption of gas species CH_4, C_2H_2, as well as H_2O in the circumstance. This spectrum is acquired with coherent averaging of around 30 seconds. The spike at around 3240 cm^-1 is an artifact signal. Inset shows a direct comparison of methane absorption in ($3000 - 3140\ cm^{-1}$) with HITRAN database.

mid-IR dual-comb interferogram is then detected by a mid-IR photodetector and recorded via an FPGA-controlled data acquisition unit that allows for coherent averaging of multiple interferogram traces.

The retrieved mid-IR frequency comb is shown in Fig. 1(c), which has a large span from $2800 - 3600\ cm^{-1}$. The spectrum features absorption of both CH_4 and C_2H_2 in the gas cell, as well as absorption of water vapour in the free-space sections of the two arms ($\sim 1.8\%$). A direct comparison with HITRAN database is also presented for these gas absorption, cf. inset in Fig. 1(c). Such a mid-IR dual-comb spectroscopy has a dynamic range of > 25 dB. The averaged signal-to-noise ratio (SNR) is $11.13/\sqrt{s}$, and the figure of merit (averaged SNR multiplying the total number of comb modes) is $\sim 10^6/\sqrt{s}$.

Discussion — In conclusion, we demonstrated a broadband mid-IR dual-comb spectroscopy for parallel gas phase detection in the range $2800 - 3600\ cm^{-1}$. The broadband coverage of the mid-IR supercontinuum, enabled by the mode hybridization in the dual-core waveguide structure, allows for parallel gas phase detection of common single-bond vibrational modes such as $C-H$, $O-H$ and $N-H$.

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