Dual-comb spectroscopy with passively mode-locked interband cascade laser frequency combs

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Abstract: We demonstrate dual comb spectroscopy using free running passively mode-locked interband cascade laser frequency combs by measuring methane around $3.6 \,\mu\text{m}$ with $100 \,\mu\text{s}$ acquisition time and an instantaneous optical bandwidth of 570 GHz.

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The interband cascade laser (ICL) is a hybrid of a conventional diode laser and a quantum cascade laser with excellent suitability for low-power spectroscopic applications in the 3-6 μ m range [1]. This was reflected in NASA's selection of ICLs for the celebrated Mars Curiosity Mission [2], which detected trace amounts of methane on Mars. Recently, this source has provided the basis for the first electrically-pumped passively mode-locked (PML) optical frequency comb (OFC) in the 3-4 μ m range, by integrating gain and saturable absorber regions on the same chip [3]. In this paper we present the first spectroscopic measurements using these novel sources.

Dual-comb spectroscopy (DCS) offers a unique potential to perform simultaneous broadband and high resolution spectroscopy in a massively-parallel fashion without any moving parts [4]. Initially, DCS was dominated by optically-pumped sources such as fiber combs, optical parametric oscillators and microresonators.



Fig. 1(a) Simplified experimental setup for the ICL-based dual-comb spectrometer. The intermode beat notes are extracted using microwave probes (photo) and bias-tees. (b) 10th harmonic of the repetition rate difference as an indicator of the timing stability. The fluctuations of the repetition rate between the combs (FWHM) are ~5 kHz over 100 µs. (c) Intermode beat note of the local oscillator comb with FWHM ~500 Hz. (d) Log-scale optical spectra of the two ICL combs. (e) Coherently averaged reverse-mapped MH spectrum acquired within 100 µs. (f) Spectroscopic measurement of pure methane at atmospheric pressure in a 10 cm absorption cell using beat notes with SNR>10 dB.

In the last few years, electrically-pumped mid-infrared Fabry-Pérot semiconductor sources emitting an output reminiscent of a FM-comb were also shown to be compatible with DCS [5,6]. In this work, we evaluate the feasibility of using PML-ICLs as comb sources for electrically-pumped DCS in the 3-4 µm range.

Figure 1(a) shows a schematic of the experimental setup. Two free running 4-mm long ICLs with gain and saturable absorber sections, as described in [3], were operated at ~15°C by injecting currents of 230 mA and 210 mA through bias-tees to the gain section of the local oscillator (LO) and signal (SIG) comb, respectively. High-numerical-aperture, AR-coated lenses collected approximately 8 mW of optical power from the output facet of each device. Biases < 3 V were sufficient to develop highly-coherent optical modes, with intermode beat note (IM BN) linewidths in the low-kHz (b) and sub-kHz (c) range. While the LO comb showed the narrowest IM BN, its optical spectrum was less uniform and sparser compared to that of the SIG comb (d). The multiheterodyne (MH) spectrum from these devices (b) covers ~23 cm⁻¹ of optical bandwidth with ~70 RF beat notes spaced by 14.8 MHz. The computationally-retrieved 10^{th} harmonic of the repetition rate difference (b) illustrates that the relative duration fluctuations of the RF interferograms are on the order of 10^{-4} , which can be compensated together with the carrier-envelope offset fluctuations, using a computational correction routine [6].

To demonstrate the spectroscopic capabilities of the PML-ICL combs, we performed 100 μ s broadband measurements of pure methane at atmospheric pressure in a 10 cm absorption cell. The results shown in Fig. 1(f) agree well with a model based on the HITRAN database, except in the region above 2780 cm⁻¹ where the LO comb exhibits low mode intensities. The short-term noise equivalent absorption of the spectroscopic measurements is in the ~10⁻³/ $\sqrt{\text{Hz}}$ range, and it will be possible to improve the optical coverage by selecting ICLs with more uniform mode structures and more closely matched IM BNs. The initial demonstration shown in Fig. 2 indicates that overlapping >30 cm⁻¹ wide combs (a) with kHz-wide IM BNs spaced by ~8.4 MHz (b) and (c) nearly doubles the optical bandwidth. Further optimization of the dispersion in the ICL cavity may also lead to broader combs with nearly uniform mode intensities spanning > 50 cm⁻¹.



Fig. 2(a) Optical spectra of the two closely matched ICL combs (vertically offset by 10 dB for clarity) and their respective kHz-wide intermode beat notes in (b) and (c) (RBW=100 Hz, VBW=1 Hz). (d) Coherently-averaged MH spectrum containing beat notes spanning 46 cm⁻¹ of optical bandwidth, of which 33 cm⁻¹ is suitable for spectroscopy on the 500 μs timescales.

In conclusion, we demonstrated dual-comb spectroscopy using a pair of free running, passively mode-locked interband cascade lasers operating at ~3.6 μ m. The optical bandwidth of ~19 cm⁻¹ (570 GHz) attainable within a sub-millisecond response time is limited mainly by an imperfect overlap of the sources, which can be improved by more careful device selection in the future. The effects of intracavity dispersion on the system performance and progress in developing a fast computational correction algorithm for free running combs will be discussed in detail.

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