Broadband high-resolution Fourier spectrometry with chip-scale combs

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***Abstract*—Electrically-pumped optical frequency combs have emerged as promising candidates for broadband, high-resolution optical sensing. Here we show how their unique emission properties can be leveraged in Fourier spectrometers with millimeter-long optical delays to obtain resolution enhancements by orders of magnitude without any optical setup modifications.**

Keywords—Fourier transform spectroscopy, frequency comb, on-chip sensing, mid-infrared, high resolution

I. Introduction

Chip-scale optical frequency combs (OFCs) [1] offer the promise of portable optical sensors for pollution monitoring and unambiguous detection of chemical warfare agents (CWA). Because CWA vary in molecular and spectral complexity, there is a demand for such coherent light sources compatible with simultaneously broadband and high-resolution spectroscopic measurements. Despite the importance, a potential difficulty arises in that many species can be detected only in the mid-infrared part of the electromagnetic spectrum [2], where the availability of comb platforms is scarcer than in the mature 1.5 μm telecom range. Fortunately, this requirement can now be addressed by self-starting OFCs generated in electrically-pumped mid-IR semiconductor lasers developed over the last decade. This emerging class of light sources is now represented by the quantum cascade laser (QCL) [3], interband cascade laser (ICL) [4], and more recently by type-I quantum well diode lasers [5] with a combined wavelength coverage of 3–10 μm, GHz line spacing, and a span of ~1 THz per device.

In principle, a pair of OFC sources can be used to perform high-resolution optical interferometry without any moving parts. The technique referred to as dual-comb spectroscopy (DCS) has gained considerable interest due to its potential for on-chip sensing at MHz rates. Multi-heterodyne optical detection encodes information from the optical domain on discrete microwave lines, each carrying information about the optical intensity and phase. However, this configuration requires high-speed photodetectors, precise microwave-bandwidth digitizers, and real-time signal processors to facilitate coherent averaging of spectroscopic data. Sharp requirements are also imposed on the OFC; pronounced amounts of optical phase noise that yield multi-MHz optical linewidths make many OFC sources incompatible with mode-resolved DCS. This is particularly severe for applications that require gap-less tunability over GHz ranges to fully resolve previously unexplored complex molecular features in the mid-infrared [6].

To address some of these limitations while maintaining a compact instrument footprint, we propose to employ the Fourier transform spectroscopy (FTS) technique enhanced by the discrete spectral pattern of chip-scale OFCs. FTS employs an optical interferometer to measure an interferogram cropped in duration due to the finite scan range of the moving arm, typically in the centimeter range. This truncation corresponds to a convolution (smoothing) in the Fourier domain, which lowers the obtainable resolution and produces ringing artifacts. Both are responsible for the notion that FTS is suitable only for coarse, less-precise optical measurements, which in turn has precluded its use in many sensing scenarios. However, fundamentally the obtainable signal-to-noise ratio in the same acquisition time is the same for both techniques while FTS requires only one laser, shows significantly more tolerance to OFC source phase noise performance, and is much easier on the signal processing side.

Maslowski et al.[7] have shown that it is possible to leverage the equidistant line spectral structure of the OFC to enhance FTS resolution by orders of magnitude. This requires matching the interferometer scan range to the source's repetition rate f_r, and nulling the offset frequency f_0 in the interferogram. The sub-nominal resolution FTS technique has rendered kHz-resolution spectroscopy (with equivalent kilometer-long optical delays) using a stabilized tabletop near-infrared OFC with multi-meter optical delay. There is, however, one significant limitation of that technique that both comb parameters (f_r, and f_0) need to be known and stabilized. Most chip-scale OFCs are incompatible with f_0 retrieval techniques like *f*-2*f* interferometry because of their narrower spectral span and lack of optical pulses required for spectral broadening. To address this challenge, we have developed a high-resolution FTS technique that requires only the knowledge of the comb's repetition rate [8], which is easily available directly from the source's bias or a photodetector.

II. METHODS AND RESULTS

In our experiment, we employ a commercial FTIR spectrometer (Bruker Vertex 80) without any modification.

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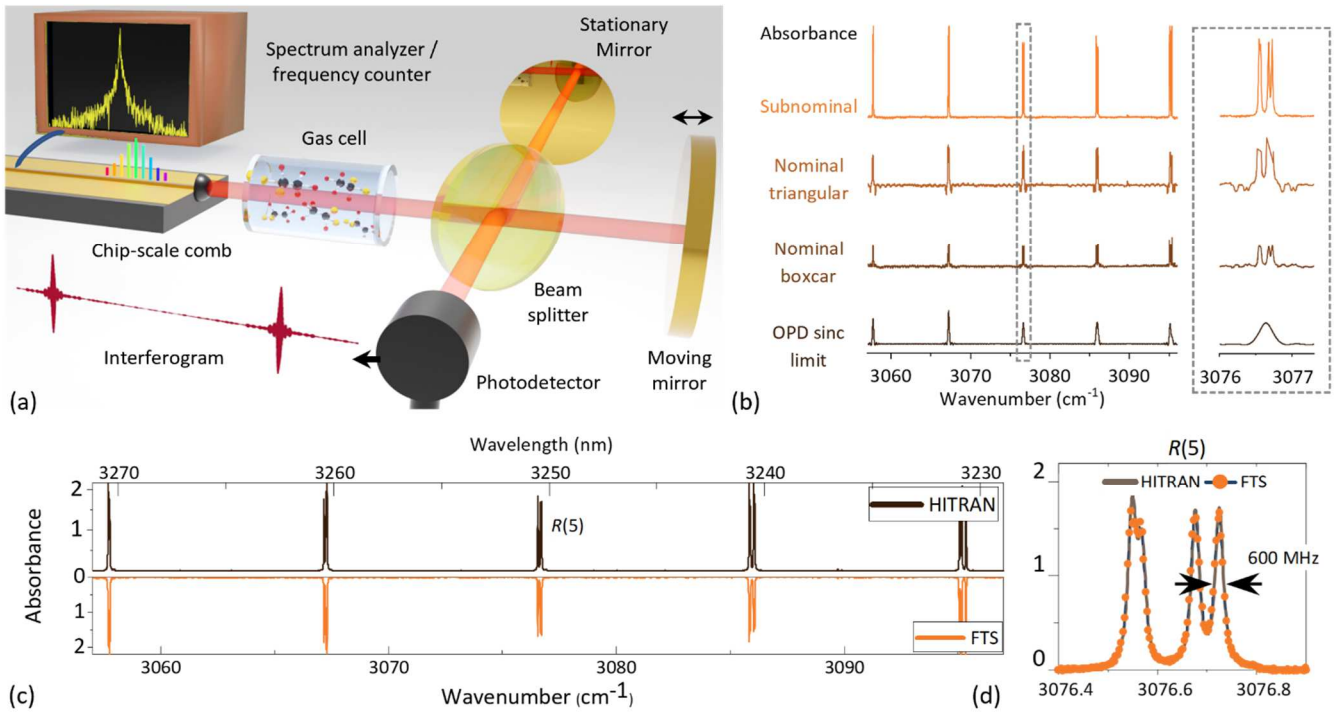

Fig. 1. Sub-nominal resolution chip-based Fourier transform spectroscopy using a 4-mm long interband cascade laser OFC and a ~10 GHz resolution Fourier transform spectrometer. (a) Experimental setup. (b) Comparison of nominal-resolution measurements of 95 Torr CH_4 using different apodization windows with the sub-nominal technique. (c) Comparision of the sub-nominal measurement with the HITRAN2020 database. (d) Zoom onto one of the *R*-branch features. Much narrower Doppler-limited absorption features can be probed at lower gas pressures [8] using complementary comb platforms.

Fig. 1a shows the simplified experimental setup, where a ~9.6 GHz repetition rate *unstabilized* ICL comb [4] operating at ~3.25 μm of wavelength is coupled into the interferometer with a nominal resolution of ~10 GHz (30 mm of optical path difference), while its repetition rate is electrically measured using a microwave spectrum analyzer. The device is step-scanned in injection current to record ~100 interleaved spectra subject to mathematical treatment described in Ref. [8]. The obtained spectra are free of distortion from the instrumental line shape (ILS), as evident from Fig. 1b. Fig.1c shows a comparison of the measured 95 Torr methane spectrum with the HITRAN database with a zoom on one of the *R*-branch feature in Fig. 1d. Excellent agreement between the FTS data and the database proves the MHz-resolution capabilities of the system despite the coarse multi-GHz nominal instrumental resolution of the FTIR.

III. OUTLOOK

Although here we use a commercial FTIR instrument, the technique can be applied to boost the resolution of custom on-chip FTS systems. It is compatible with virtually any comb emitter in any spectral region without any setup modifications, and has a great potential to find application in battery-operated spectrometers in terrestrial and space applications.

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