MHz-Resolution Fourier Transform Spectroscopy with Millimeter-Scale Optical Path Differences

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Abstract: Enhancing the resolution of a Fourier transform spectrometer several hundred times to a MHz level is presented. A chip-scale optical frequency comb as a source enables high-resolution spectroscopy without any changes to the optical setup.

1. Introduction

Over the past several decades, Fourier transform spectroscopy (FTS) has established its position as a key technique for the characterization of absorption spectra of gaseous, liquid, and solid analytes. By interfering light with its delayed copy on a photodetector, in FTS one obtains a field autocorrelation signal whose Fourier transform yields an optical spectrum, or more precisely, its power spectral density. Typical experimental realizations comprise a Michelson interferometer, as shown in Fig. 1a.

Whereas the FTS technique has the main advantage of compatibility with any light source, including broadband incoherent thermal emitters, it suffers from two major issues relating to a finite scan range of the optical delay line. Truncation of the FTS signal referred to as the interferogram convolves the measured spectrum with an instrumental line shape (ILS), which in turn lowers the obtainable resolution to approximately $1/\Delta$ [cm^{-1}], where Δ is the optical path difference (OPD). Typical commercial-grade FTS instruments offer ~GHz resolution with cm-long OPDs. Unfortunately, for precise spectroscopic investigations, it is often insufficient. The second issue relates to the fact that the convolution induces ringing artifacts due to the wings of the sinc function visible as spectral artifacts. Maslowski et al. have proposed a sub-nominal resolution FTS technique to suppress the influence of the ILS if one leverages the unique, discrete structure of an optical frequency comb [1]. Stabilization of the comb's offset and repetition rate frequency permits high-fidelity spectroscopic measurements with resolution in the kHz range. However, because of the source's low repetition rate (100~MHz), custom-made, meter-long OPD spectrometers have to be used. This large footprint precludes a widespread adoption of the technique in more portable scenarios.

To address this issue, we propose a sub-nominal FTS technique that lifts the requirement of offset frequency stabilization or even its knowledge [2]. We extract it directly from the measured interferogram provided one knows the repetition rate, which is easily retrievable from electrical bias or photodetector. This unlocks the high-resolution potential of chip-based optical frequency comb platforms [3], whose offset frequency cannot be retrieved via *f*-to-2*f* interferometry. Also, because of the GHz repetition rate offered by such devices, mm-long OPDs are sufficient.

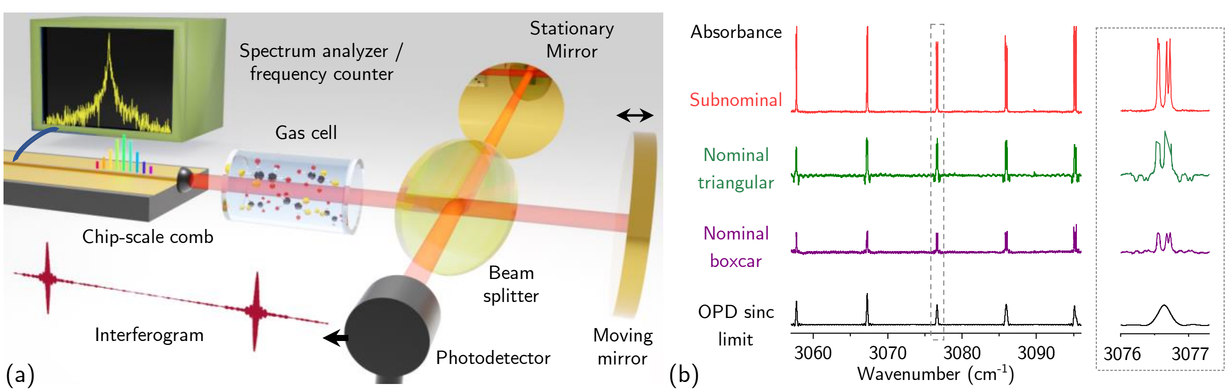

Fig. 1. Sub-nominal resolution Fourier transform spectroscopy (FTS) using an interferometer with a mm-long optical delay line. (a) Experimental setup. (b) Comparison of conventional FTS measurements of low-pressure methane in a natural isotopic ratio (95 Torr) with the sub-nominal FTS routine. The nominal resolution traces are calculated by extracting peak intensities from mode-resolved intensity spectra (interferograms capture two bursts). In all cases, except for the sub-nominal case, ringing artifacts, peak rounding, and negative absorbance appear.

2. Results

To perform sub-nominal FTS, we use a Bruker Vertex 80 FTIR spectrometer without any modifications. Light from a chip-scale tunable optical frequency comb with electrical pumping is coupled into the instrument and next, single-sided interferograms are recorded. Fig. 1b already shows that extraction of the peak intensity from nominal-resolution, mode-resolved spectra is not sufficient to obtain quality spectroscopic data. A more consistent spectroscopic investigation is shown in Fig. 2. Spectrally-interleaved measurements enable MHz-resolution spectroscopy using an instrument with a nominal resolution of 100 MHz. Panels 2a–b show the absorption spectrum of methane measured using an interband cascade laser frequency comb [4], while 2c–d illustrate the measurement of low-pressure acetylene measured using a diode laser frequency comb [5]. In all cases, excellent agreement between the HITRAN2020 database [6] and FTS data is obtained.

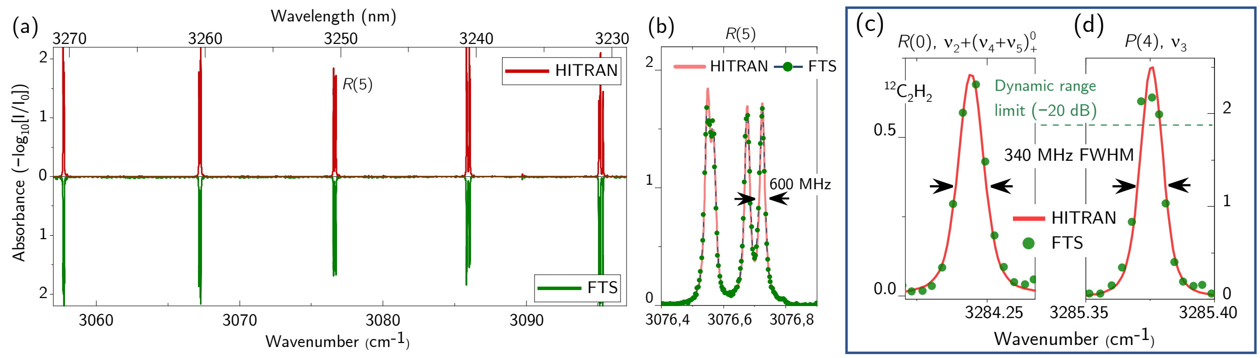

Fig. 2. Sub-nominal resolution Fourier Transform spectroscopy using ~10 GHz repetition rate tunable optical frequency combs. (a) The ν_3 mid-infrared band of methane (CH_4) probed by a 4-mm long interband cascade laser frequency comb [4] compared with the HITRAN2020 database [6]. The analyte pressure was 95 Torr. (b) Zoom onto one of the *R*-branch features. (c, d) Measurement of acetylene $^{12}C_2H_2$ at 10 Torr using a diode laser frequency comb at 3 μm [5]. Resolving such narrow, Doppler-limited transitions practically requires an instrumental resolution better than 100 MHz.

3. Conclusion and outlook

We present a versatile technique to perform high-resolution Fourier spectrometry using chip-scale optical frequency comb sources, whose offset frequency cannot be retrieved using conventional *f*-to-2*f* interferometry. Because the technique requires only several frequency estimation steps and careful computation of the frequency spectrum, it can be easily applied to boost the resolution of existing FTIR instruments if the light source has frequency comb properties. Compact on-chip FTS instruments are also expected to benefit from our technique if one utilizes frequency-tunable devices with repetition rates on the order of 10's of GHz. In the future, piezo-actuated linear stages should allow for the realization of extremely compact instruments with space-compatibility in even challenging regions like the THz.

Acknowledgment: This work was supported under National Aeronautics and Space Agency's (NASA) PICASSO program (106822 / 811073.02.24.01.85), and Research and Technology Development Spontaneous Concept Fund. It was in part performed at the Jet Propulsion Laboratory (JPL), California Institute of Technology, under contract with the NASA. L. A. Sterczewski's research was supported by an appointment to the NASA Postdoctoral Program at JPL, administered by Universities Space Research Association under contract with NASA. L. A. Sterczewski acknowledges funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 101027721. The authors would like to thank Dr. Kevin Lascola, and Dr. Feng Xie at Thorlabs Inc. for providing the ICL material, and Dr. Clifford Frez at JPL for providing the diode laser material used in this study. Dr. Jerry Meyer, and Dr. Igor Vurgaftman at NRL are acknowledged for fruitful discussions on ICL combs.

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