

Breaking the Delay-Resolution Limit of Fourier Transform Spectrometers Using Chip-Scale Combs

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Abstract: Resolution enhancement of Fourier transform spectrometers from GHz to MHz level is demonstrated without any changes to the optical setup. The technique leverages the discrete spectral structure of a chip-scale optical frequency comb. © 2023 The Authors

1. Introduction

The Fourier transform spectrometer is arguably one of the most popular optical instruments with applications in diverse areas of science. It plays a key role in the characterization of gaseous, liquid, or solid samples. A Michelson- or Mach-Zehnder-type interferometer measures the electric field autocorrelation, whose Fourier transform yields the optical spectrum. Compatibility with virtually any optical source, including incoherent thermal emitters, makes this instrument of preference for measurements in any spectral range. Unfortunately, the Fourier transform spectroscopy (FTS) technique exploiting an arbitrary light emitter suffers from the delay-resolution limit. Mechanical displacement of Δ (in cm) dictates a spectral resolution of $1/\Delta$ (in cm^{-1}). In practical realizations, centimeter-long delays offer resolutions in the multi-GHz range, which are insufficient for simple gaseous analytes, particularly at lower pressures. Another concern in FTS is a convolution of the measured spectrum with an instrumental line shape function (ILS) due to the finite optical displacement. Not only does it wash out narrow spectral features, but also produces ringing spectral artifacts. Maslowski et al. have proposed to leverage the discrete spectral structure of a stabilized optical frequency comb with MHz repetition rate (f_{rep}) to perform FTS with ILS suppression [1]. The technique that requires measuring the interferogram over exactly c/f_{rep} offers resolution in the kHz range or even better with corresponding displacements in the kilometer range. However, the source's relatively low repetition rate dictates the use of custom meter-long delay lines, which merged with the knowledge and stabilization of the comb's offset frequency f_0 via f -to- $2f$ interferometry limits its applicability to a laboratory environment. To circumvent these shortcomings, we propose a sub-nominal resolution FTS technique that lifts the f_0 knowledge or stabilization requirement [2]. Instead, f_0 is extracted directly from the measured interferogram provided the laser repetition rate f_{rep} is known, which unlike f_0 is easy to obtain directly from the photodetector or device bias. By using unstabilized chip-scale electrically-pumped frequency combs with GHz repetition rates and electrical tunability [3], we perform MHz-resolution spectrometry of gaseous samples at low pressure using only millimeters of optical displacement. This corresponds to a resolution enhancement by almost 3 orders of magnitude. Note that in the mid-infrared range, where our sources operate, f_0 retrieval using conventional techniques would be impractical because the field waveform has a nearly constant intensity (i.e. frequency-modulated) instead of being of pulsed nature [4,5].

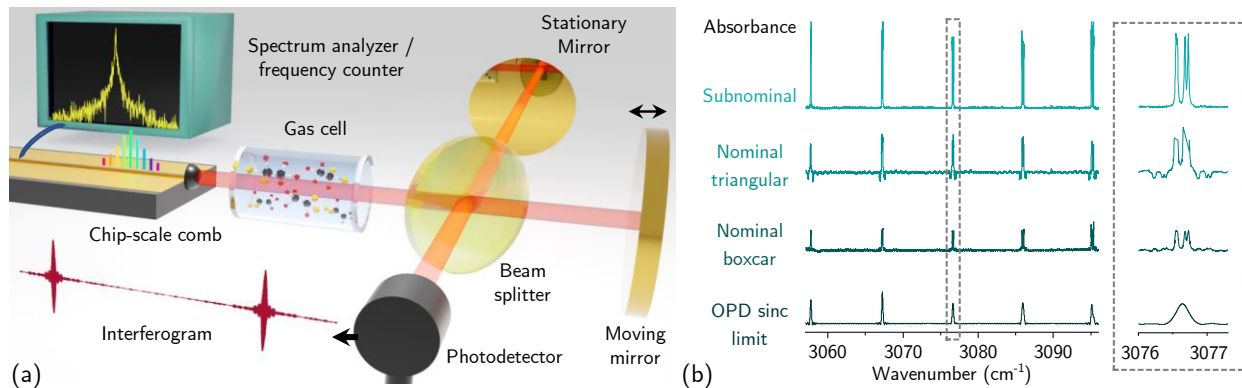


Fig. 1. (a) Experimental setup of the sub-nominal FTS technique. (b) Comparison of different techniques for calculating the absorption spectrum of low-pressure methane. Peak value extraction fails to provide quality spectroscopic data. Only the sub-nominal resolution routine correctly resolves the complex features of CH_4 .

2. Results

The experimental setup shown in Fig. 1a corresponds to a commercial Bruker Vertex 80 FTIR spectrometer without any modifications. An interband cascade laser frequency comb at 3.25 μm [4], and a diode-laser-based 3.05 μm comb [5] are used for measuring methane, and acetylene, respectively. The devices are tuned in injection current for spectral interleaving. Fig. 1b shows that simple peak extraction of mode-resolved spectra acquired from two-burst interferograms is not sufficient to faithfully reproduce narrow absorption features of low-pressure methane (CH_4) in the ν_3 mid-infrared band. In contrast, the sub-nominal resolution routine described in detail in Ref. [2] provides high-fidelity data of 600-MHz wide lines when the nominal spectrometer resolution is 9.6 GHz. Even more demanding absorbers like acetylene ($^{12}\text{C}_2\text{H}_2$ at 10 Torr) can be probed by the 3.05 μm diode combs, as shown in Fig. 2. At such low pressures, the absorption linewidth is only ~ 340 MHz, which practically requires an instrumental resolution better than 100 MHz to faithfully reproduce its shape (3 meters of displacement in FTS). A comparison with the HITRAN2020 database [6] confirms excellent agreement between the measured sub-nominal resolution, and modeled spectra except for weak fringe-like optical feedback noise which does not plague the ICL-based measurement [2]. Figures 2(b) and 2(c) zoom onto two absorption lines with different strengths.

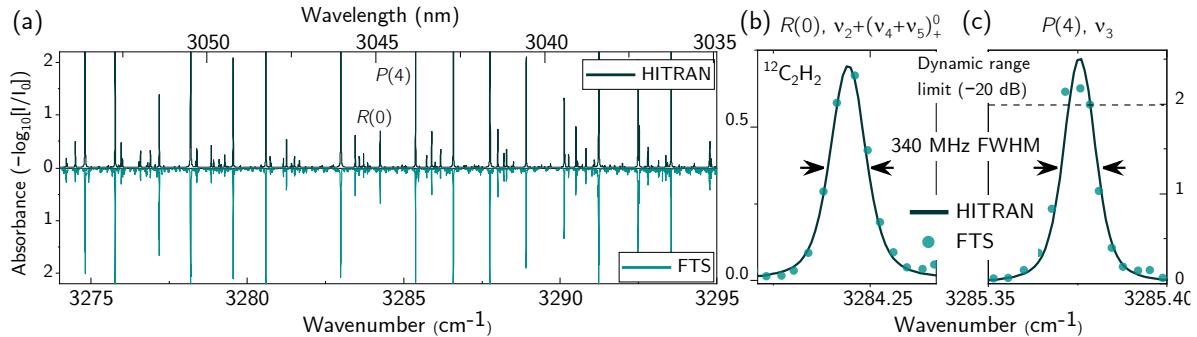


Fig. 2. Sub-nominal resolution Fourier Transform spectroscopy using a ~ 10 GHz repetition rate tunable optical frequency comb. (a) Measurement of acetylene $^{12}\text{C}_2\text{H}_2$ at 10 Torr using a diode laser frequency comb at 3 μm . Resolving narrow, Doppler-limited transitions (b,c) practically requires an instrumental resolution better than 100 MHz (≥ 3 m of optical delay), while the spectrometer in this experiment offers 10 GHz.

3. Conclusion and outlook

We present a versatile sub-nominal resolution Fourier transform spectroscopic technique that can be readily used to boost the resolution of existing FTIR instruments almost a thousand times. Unlike in the case of the dual-comb spectroscopy technique, the optical linewidth constraints are significantly relaxed, which permits the use of less mature comb sources with pronounced amounts of phase noise for demanding spectroscopic measurements in any spectral region. Beyond commercial instruments, this technique offers potential for the realization of broadband, chip-scale piezo-driven spectrometers with battery operation capabilities. Virtually any free-running, tunable source with frequency comb properties and preferentially higher (GHz) repetition rate should serve this purpose.

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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