1 GHz mid-infrared dual-comb spectrometer spanning more than 30 THz

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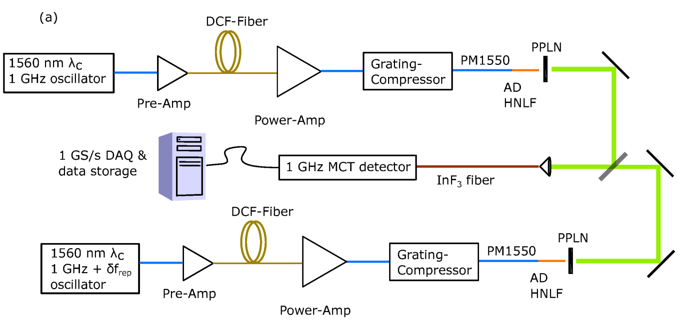
Abstract: We demonstrate a broadband 1 GHz mid-infrared dual comb spectrometer based on intra-pulse difference frequency generation, addressing the 3 – 5 μm wavelength region for the characterization of dynamic molecular systems.

1. Introduction

Simultaneous identification of multiple molecular species in dynamic systems is enabled by mid-infrared (MIR) spectrometers that occupy the intersection of three metrics: high speed, high frequency resolution, and broad bandwidth. High resolution and bandwidth in the region of fundamental absorption benefit the identification of multiple molecular species, and fast spectral acquisition is of particular interest for systems that evolve on short, e.g. sub-millisecond, time scales.

A powerful approach for infrared high speed spectroscopy is the dual comb spectrometer (DCS), which combines high frequency resolution and spectral acquisition rates orders of magnitude higher than those of conventional Fourier-transform spectroscopy. This technique has matured in the telecom wavelength region where compact lasers, amplifiers and fiber components are available [1]. In the MIR, platforms based on nonlinear down conversion of near-infrared combs have generated bandwidths covering multiple octaves [2, 3], but at repetition rates on the order of one hundred megahertz. With such comb spacing, the tradeoff between spectral coverage and spectral acquisition rate has limited MIR DCS to detection bandwidths less than $\sim$ 20 THz when data acquisition rates exceed $\sim$ 1 kHz (with numbers given for 200 MHz mode spacing) [4, 5]. Characterization of systems with multiple molecular species that evolve on the tens to hundred microsecond time scales would benefit from MIR frequency combs that combine broad spectral coverage ($>$ 30 THz) with operation in the gigahertz regime. To address these challenges, we demonstrate a 1 GHz broadband MIR dual comb spectrometer that is based on a simple and yet robust single-branch intra-pulse difference frequency generation (IP-DFG) platform.



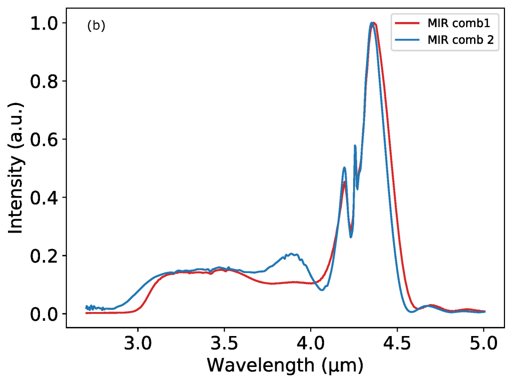


Fig. 1. One gigahertz MIR dual comb spectrometer (a) Schematic of MIR generation and detection. (b) Optical spectra from the two frequency combs.

2. Experiment and Results

The experimental setup is sketched in Fig. 1 (a). Two fully stabilized 1 GHz turnkey oscillators centered at 1560 nm are amplified to roughly 4W of power in a fiber amplifier. Each consists of a nonlinear core-pumped erbium doped fiber pre-amplifier, several meters of dispersion compensating fiber that acts as a pulse stretcher, a cladding pumped erbium/ytterbium doped power-amplifier, and a free-space grating pair for temporal recompression. The output is recoupled into a polarization maintaining (PM) fiber patch cord that is terminated by a few centimeters of PM anomalous dispersion highly nonlinear fiber (AD-HNLF). The length of HNLF is designed to meet the pulse at its fission point to obtain optimal bandwidth ($\sim 1-2\ \mu$m) and a pulse with duration as short as 8 fs. This few-cycle pulse is focused into a 1mm thick fanout periodically poled lithium niobate crystal (PPLN), generating up to 3 – 4 mW of light within $3-5\ \mu$m via intrapulse difference frequency generation. An example of the broadest achievable spectra is shown in Fig. 1(b), consisting of 33,310 discrete comb teeth across 33 THz (1100 cm^{-1}).

The outputs of the two mid-infrared frequency combs are combined at a beam-splitter and mode-matched by coupling the combined beams into indium-fluoride fiber. The heterodyne beat is detected on a high-speed HgCdTe mid-infrared detector, whose analog output is digitized at 1 GHz using a field programmable gate array (FPGA). The FPGA continuously streams the digitized samples concurrently with data acquisition to computer RAM.

The single-shot and averaged DCS spectra (full 1 GHz resolution with 19,900 spectral elements) for a ten percent methane mixture at 600 Torr inside a 10 cm cell are plotted in Fig. 2 (a), with CO_2 absorption also evident around 4.2 μm. Cascaded $\chi^{(2)}$ nonlinearities simultanesously provides access to the offset frequencies of the near-infrared combs, which can be employed for frequency stabilization [6]. The present signal-to-noise ratio (SNR) is limited by laser RIN. Fig. 2 (b) shows the absorbance noise with 1 GHz resolution, which improves with the square-root of averaging time. For high-speed gas-phase spectroscopy, the metric for SNR will be the accuracy with which one can retrieve relevant thermodynamic parameters via models that are fit to the absorption features. The broad bandwidths we achieve will be beneficial in this analysis, as the uncertainty of the data fitting improves with the number of absorption features.

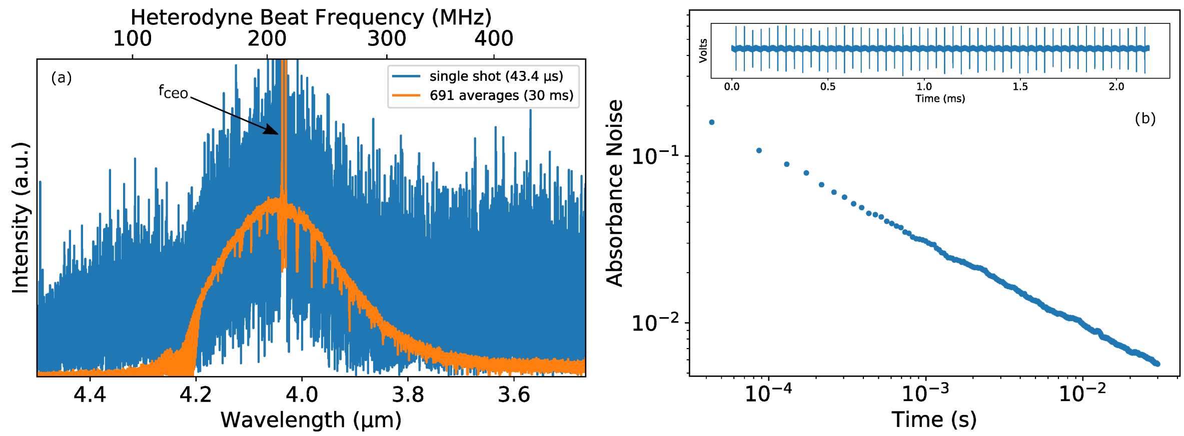


Fig. 2. Dual comb spectra and noise (a) Single-shot DCS spectrum with 1 GHz resolution, and the same spectrum after 30ms of averaging time. (b) Absorbance noise as a function of averaging time (1 GHz resolution). (Inset) Acquistion of 50 interferograms over 2 ms.

In conclusion, we demonstrate a 1 GHz mid-infrared dual comb spectrometer with >30 THz spectral coverage in the $3-5\ \mu$m region. We anticipate that the combination of extensive MIR bandwidth, spectral resolution and high-speed acquisition will prove useful in characterizing dynamic and complex molecular systems in a wide range of physical, chemical and biological applications.

References

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