Dual Chirped-Pulse Electro-Optic Comb Generation in the THz Region: Spanning the Spectroscopic and Quantum Dynamics Domains

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Abstract: A new THz difference-frequency chirped-pulse dual-comb method based on electro-optic phase modulators (EOMs) and two near-visible continuous wave lasers is used to perform high resolution spectroscopy and to magnify the rapid passage signal response. © 2022 The Author(s) Official contribution of the National Institute of Standards and Technology; not subject to copyright in the United States.

1. Introduction

Dual optical frequency combs (DOFC) have proven to be a versatile tool for precision metrology and sensitive spectroscopic sensing. One key advantage of the method is the spectral bandwidth compression in down-conversion and the unique mapping of the comb teeth in the optical domain to the radiofrequency (RF) region for high-throughput detection [1,2]. In photon starved applications where limited spectral coverage is desired to enhance sensitivity, electro-optic DOFCs (EO-DOFC) have provided a facile route to optimize the comb resolution and bandwidth. In recent work using a single free-running near-IR laser [3], we have shown a further advantage can be realized using dual chirped-pulse EO-DOFCs to temporally magnify the quantum dynamics of molecular systems. A magnified view of the rapid passage complex signal response (magnitude and phase) of CO_2 was observed by applying a differential chirp rate between the signal (SIG) and local oscillator (LO) legs of the interferometer.

In this work, the quantum dynamics of water vapor are investigated in the THz region over a range of pressures that transform the normal complex line shape to the rapid passage signal response. The pure rotational line of water, $1_{1,0} \leftarrow 1_{0,1}$ ($J_{Ka,Kc}$ notation), is investigated at 556.936 GHz (18.577 cm^{-1}). For a 10 ms scan, the temporal magnification factor realized in this work is 250,000. This is more than 1600-fold larger than that demonstrated in the previous near-IR study [3]. The observed response is modeled using the Maxwell-Bloch Equations (MBE) [4].

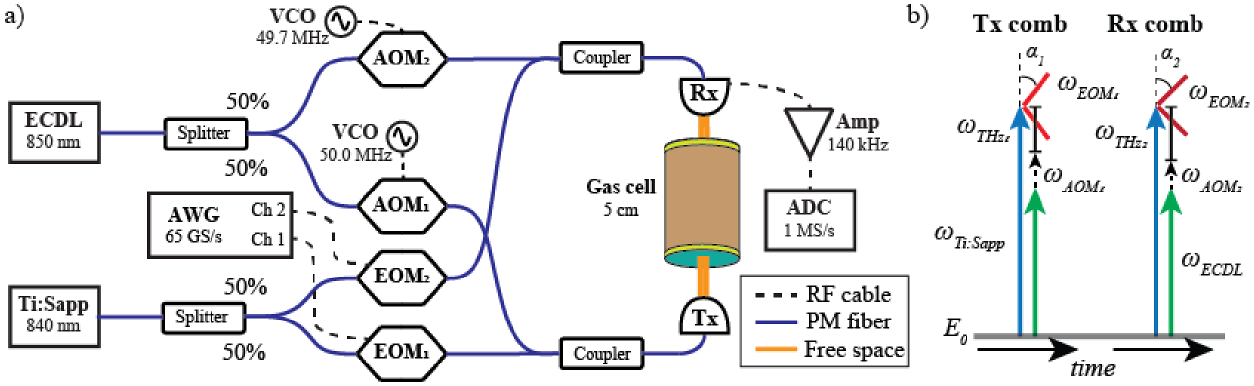

Figure 1: a) Schematic of the interleaved DF-EO-DOFC spectrometer. b) A schematic illustration of the difference frequency system.

2. Experiment

The difference-frequency electro-optic dual-optical-frequency-comb (DF-EO-DOFC) system is shown in Fig. 1a. The system consists of two continuous wave (CW) lasers, an external cavity diode laser (ECDL) and a Ti:Sapphire (Ti:Sapp) ring laser. The two lasers are coupled into polarization maintaining fibers (120 mW in each) and then split into two equal legs. The ECDL outputs are passed through two acoustic-optic modulators (AOMs) that operate near 50 MHz. The Ti:Sapp outputs are coupled to two EOMs, with each EOM driven by a channel of a fast (32 GS/s) arbitrary waveform generator. Each EOM output is combined (50% / 50%) with one of the AOM outputs to define the transmitter (Tx) and receiver (Rx) legs. The combined outputs are fiber coupled to a photomixer transmitter that emits mildly focused THz radiation through a 5 cm evacuable cell to a photomixer receiver.

The scheme for THz wave generation is illustrated in Fig. 1b. The ECDL and Ti:Sapp laser frequencies are represented as vertical green and blue lines, respectively. The AOM offset frequencies are shown as dashed black lines and the EOM ranges covered by the chirped pulses at chirp rates, α_1 and α_2, are shown as tilted red lines for the

(±) sidebands. The resulting THz frequency ranges generated on the Rx and Tx are indicated with solid black lines. The RF comb detected is generated via mixing of the optical and THz waves on the Rx and digitized at a rate of 1 MHz over a total period of 1 sec. The widths of the RF comb teeth are < 1 Hz.

The two EOMs on the Tx and Rx legs are driven with chirped pulse waveforms defined according to the following,

$$WF_i^{Tx}(t) = sin\left(2\pi f_{Tx_{start}} t + \frac{2\pi(fTx_{stop} - fTx_{start})}{2\tau_{CP}} t^2\right) \tag{1}$$

$$WF_i^{Rx}(t) = sin\left(2\pi f_{Rx_{start}} t + \frac{2\pi(fRx_{stop} - fRx_{start})}{2\tau_{CP}} t^2 - \frac{2\pi}{N_{chirps}}\left(\frac{t}{\tau_{CP}} + i\right)\right) \tag{2}$$

$$i = 1 \ldots N_{chirps}, \quad t = 0 \ldots \tau_{CP}$$

where τ_{CP} is the chirp duration that defines the RF comb resolution (100 Hz) and N_{chirps} defines a frequency shift in the RF region that depends on the EOM order (10 Hz).

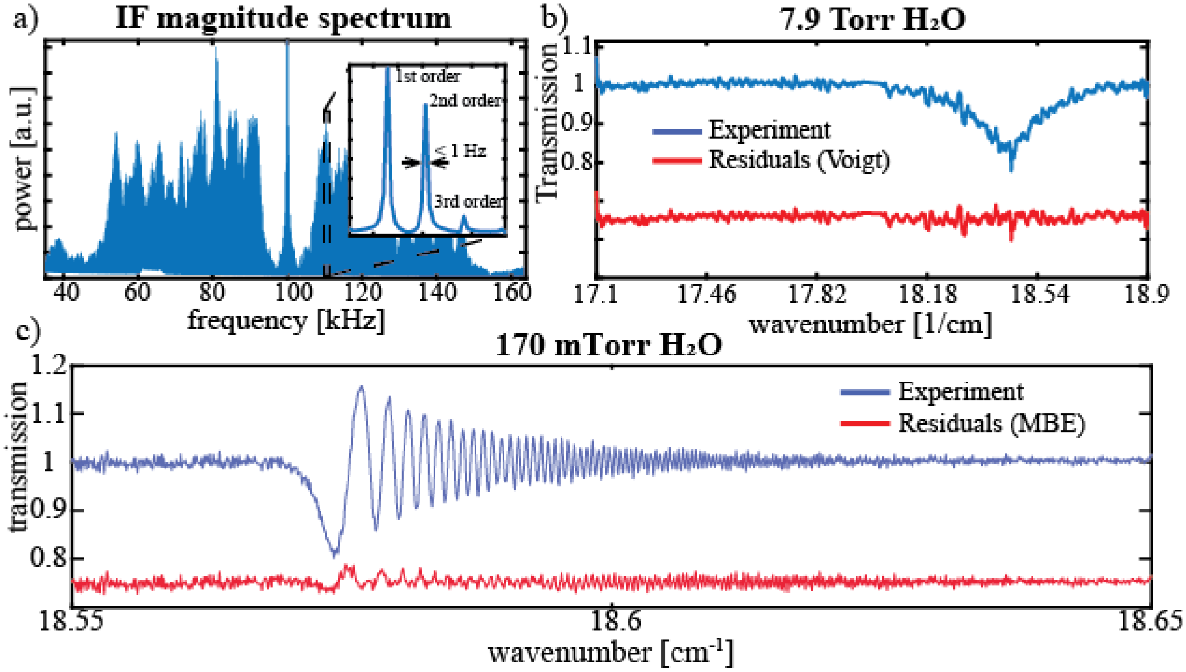

Figure 2: a) The RF comb spectrum centered at 100 kHz, showing the (+)/(-) comb teeth on the right/left side of the center. The inset shows the interleaving of the different orders up to 3rd-order and the Hz level widths of the comb teeth. b) The transmission (magnitude) spectrum of the $1_{1,0} \leftarrow 1_{0,1}$ ($J_{,Ka,Kc}$ notation) transition of H_2O at 556.936 GHz (18.577 cm^{-1}) at a pressure of 7.9 Torr (1.0 kPa) in about 640 Torr (85 kPa) of N_2. The residuals from a Voight fit are shown below in red. c) The observed rapid response of H_2O vapor at a pressure of 170 mTorr. The residual determined from the fit of the data to the MBE model are shown below in red. The temporal magnification of the response is 250,000.

3. Results

The DF-EO-DOFC method is used to measure the pure rotational line of H_2O in the THz region using lasers operating near 842 nm. Figure 2a shows how the start and stop frequencies of 0.5 GHz and 15 GHz for f_{Tx} that span 30 GHz of optical bandwidth are mapped down to the RF region from 10 kHz to 50 kHz in the second order. The transmission (magnitude) data shown in Figs. 2b and 2c were obtained for the $1_{1,0} \leftarrow 1_{0,1}$ transition of H_2O. A normal Voigt line shape spectrum is obtained at near-atmospheric pressure (1 kPa of H_2O in 82 kPa of N_2). However, at low pressure such as in Fig 2c, the spectrum is seen to transform to a rapid passage signal response with quantum dynamics that extend over a large part of the 11 GHz spectrum (only 3 GHz is shown for the (+) sideband). The extreme change in the spectral response is a result of the large temporal magnification of 250,000 as determined from the ratio of the optical and RF bandwidths, $\alpha = \Delta f_{Opt}/\Delta f_{RF}$. As evident from the residuals (in red), the response is well-modeled using the MBEs. Further studies of temporal magnification are underway for other gas phase molecules and condensed phase systems.

4. References

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