

# Rapid Passage Signals from CO<sub>2</sub> at 1.6 μm Using a Dual Chirped-Pulse Electro-Optic Comb System with High-Order Interleaving

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**Abstract:** We demonstrate a method for interleaving high-order sidebands in a fast-sweeping electro-optic dual-comb system. We probe CO<sub>2</sub> at 1.6 μm and show sweep-rate dependent rapid-passage effects modeled with the Maxwell-Bloch equations. © 2021 The Author(s)

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## 1. Introduction

Over the past decade, optical frequency combs have revolutionized spectroscopy by providing high-resolution spectral sampling and excellent phase-noise stability. Further, electro-optic based comb systems can use a single laser source to seed multiple combs, simplifying the optical setup and sources needed for dual-comb spectroscopy [1]. The down-conversion of optical frequencies into radio frequencies (RF) in dual-comb systems is determined by the difference in the repetition rate, or tooth spacing, between the two combs. These dual comb techniques have been shown in a variety of formats for gas phase reaction dynamics and line shape studies [2,3].

We present a dual-chirped electro-optic comb system where we take advantage of the high orders of the electro-optic modulator (EOM) to increase the effective optical bandwidth with electronic bandwidth limited sources. As opposed to 1<sup>st</sup>-order scanning using low power microwave sources to avoid interference from the higher order EOM modes [4], we separate both the high order tones as well as the positive and negative sidebands using acousto-optic modulators (AOM). In this paper, we present a flexible, fast-sweeping dual-comb spectrometer using electro-optic modulators driven by an arbitrary waveform generator (AWG). We observe large rapid passage signals of CO<sub>2</sub> absorption lines over a 120 GHz (4 cm<sup>-1</sup>) region near 1.6 μm and compare our results to predictions from the Maxwell-Bloch equations [5].

## 2. Experiment

The system shown in Fig. 1 uses a single, tunable external cavity diode laser (ECDL) that is actively locked to an external cavity with a short-term stability of  $\pm 250$  kHz. The laser output is split into three legs: a lock leg, local oscillator (LO), and signal leg. Using AOMs, the lock leg is shifted by -350 MHz, and the LO and signal legs are shifted 50.000 MHz and 49.979 MHz, respectively, to generate beat notes at 400.000 MHz and 399.979 MHz on a balanced APD detector. The two beat notes are phase locked to external RF sources to stabilize the 21 kHz beat note between the signal and LO legs that serves to separate the +/- sidebands. A two-channel 65 GS/s AWG drives the two EOMs and is programmed with tailored chirped waveforms to control the scan time, frequency range and spectral resolution of the system. The waveforms driving the EOM of the LO leg can be written as:

$$WF_i^{LO} = \sin\left(2\pi\left(f_{start} - f_{IFstart} - \frac{i}{N_{chirps_t}}\right)t + \frac{(f_{stop} - f_{start} - f_{IFstop})}{2\tau_{CP}}t^2\right) \quad (1)$$

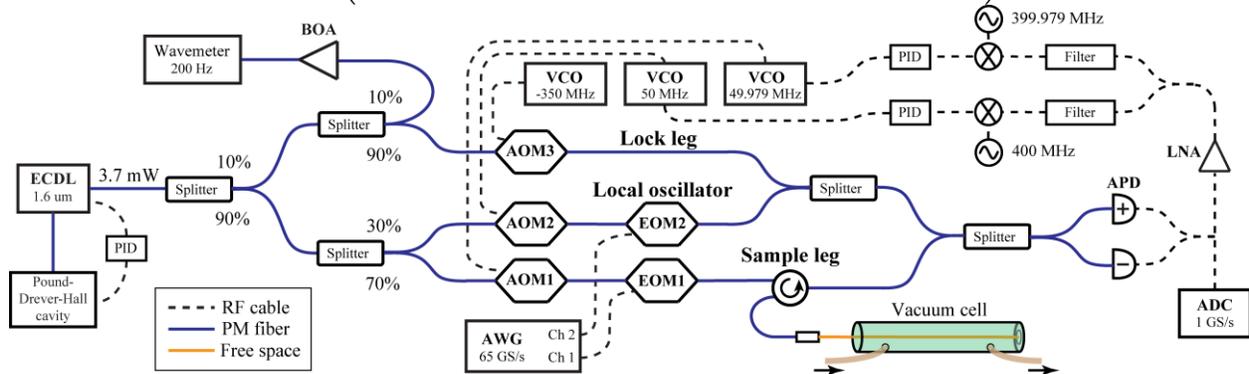


Fig. 1. The dual chirped-pulse electro-optic comb system equipped with a 1.12 m double-pass cell. External cavity diode laser (ECDL), proportional-integral-differential control (PID), booster optical amplifier (BOA), acousto-optic modulator (AOM), electro-optic modulator (EOM), arbitrary waveform generator (AWG),

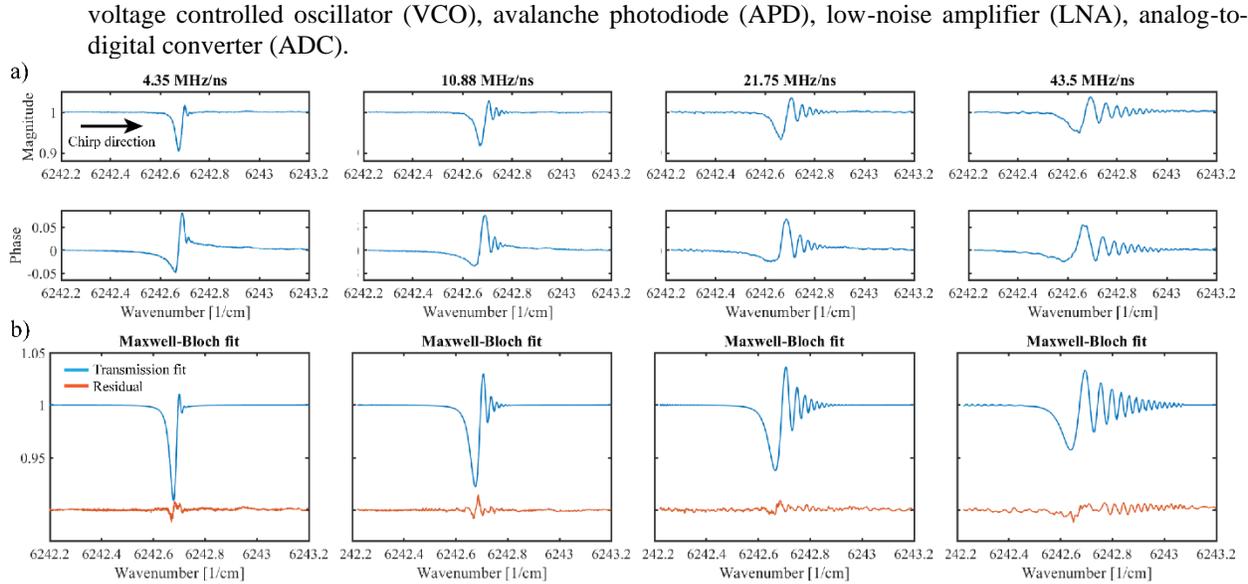


Fig. 2. Third order sideband CO<sub>2</sub> transmission line at 6242.6721 cm<sup>-1</sup> and 6.6 kPa. a) The lines from left to right show an increase in the relaxation duration as the sweep rate is increased from 4.35, 10.88, 21.75, and 43.5 MHz/ns. b) The Maxwell-Bloch fit and residuals, to scale, to our experimental data shows the excellent agreement with the two-level model.

where the scan range of the first order is  $f_{start}$  to  $f_{stop}$ , the chirp duration is  $\tau_{CP}$ , and the bandwidth of the down-converted signal is  $f_{IFstart}$  to  $f_{IFstop}$ . The signal leg EOM is chirped from  $f_{start}$  to  $f_{stop}$ , while the LO iterates through  $i = 1 \dots N^{chirps}$ , introducing a phase slip that separates each of the higher orders of the EOM by  $m/(\tau_{CP}N^{chirps})$ , where  $m = 1, 2, 3$ , etc., is the order. These waveforms have a repetition rate equal to  $1/\tau_{CP}$  and generate chirped interference patterns, which when Fourier transformed produce comb lines in the RF spectrum with that spacing. We scan the 1<sup>st</sup>-order frequency range from 500 MHz to 15 GHz with the AWG to generate comb lines in the EOMs up to 4<sup>th</sup> order and equalize the 4<sup>th</sup>-order intensities from  $\pm 2$  GHz to  $\pm 60$  GHz.

The signal leg is double passed through a 1.15 m cell containing CO<sub>2</sub> at 6.6 kPa (50 torr). The collimated beam has a  $\approx 1$  mm beam waist and  $\approx 0.2$  mW of average power. The circulated light is mixed with the combined output from the LO and lock legs to generate interferograms on a 500 MHz balanced detector. To recover the transmission spectra of CO<sub>2</sub>, we Fourier transform the detected signal and sample at the comb teeth generated by the repeating frequency chirp.

### 3. Results

The CO<sub>2</sub> line investigated here is at 6242.6721 cm<sup>-1</sup> and has a line strength of  $\approx 1 \times 10^{-23}$  cm<sup>-1</sup>/(molecule cm<sup>-2</sup>). We can tune the sweep rate from 4.35 MHz/ns to 43.5 MHz/ns, by adjusting the chirp duration  $\tau_{CP}$  from 10  $\mu$ s to 1  $\mu$ s. In figure 2a, we show the transmission and phase spectra for four different sweep rates of the 3<sup>rd</sup> order sidebands. We can see a clear frequency and intensity dependence of the ripple on the sweep rate. At low sweep rate, the line profile is nearly symmetric, showing a small, high frequency ripple on the upper end of the line. As the chirp time decreases, we see significant increases in the slowly oscillating, rapid passage signals in both the phase and amplitude spectra.

In prior work with quantum cascade lasers [5], the Maxwell-Bloch equations (MBE) have been used to model rapid passage effects for two level systems. The MBE fits to our transmission profiles are shown in Fig. 2b, along with the residuals, indicating we are indeed seeing the effects of relaxation of the Feynman-Bloch vector, resulting in the observed coherent emission ripples. This chirped-pulse comb system provides the scan precision and flexibility to gain valuable insight into the physics of the high-speed and low-pressure relaxation dynamics of molecular systems and is currently being adapted to advance remote sensing techniques for greenhouse gases.

### 4. References

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