Multiheterodyne Spectroscopy Using Multi-frequency Combs

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Abstract: Near-IR dual frequency combs generated from waveform driven electro-optic phase modulators (EOMs) are used for high resolution studies in low pressure cells and for remote sensing from natural targets (Boulder Flatirons). Arbitrary waveform generators (with up to 20 GHz of bandwidth) enable amplitude (AM), phase (PM) and harmonic (HM) modulation control of dual parallel phase modulators to generate tailored frequency combs for sensing requirements.
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Frequency combs generated using radiofrequency(rf)-driven electro-optic phase modulators have proved advantageous in a wide variety of areas from low pressure gas cell studies [1] to sub-Doppler spectroscopy [2]. Dual comb methods [3-7] are particularly valuable to enable high throughput collection over wide spectral regions that exceed detector bandwidths for a single comb. Dual parallel Mach-Zehnder phase modulators (MZM) driven with arbitrary waveform generators (AWGs) bring additional real time single element control of the interferogram to optimize comb properties. Comb tooth resolution/interferogram scan rate, optical bandwidth and relative phase can be readily adapted to sensing requirements while maintaining optimal conditions for data throughput.

In this work, we generate frequency combs using MZMs driven by AWGs that originate from a single external cavity diode laser tunable over a range from 1600 nm to 1646 nm. The basic elements of the system are shown in Fig. 1 (left) [6]. For the gas cell experiment, the output of the ECDL is fiber split to generate combs in two MZMs for the probe (+reference) and local oscillator (LO) arms. The signal arm is split again and separate frequency offsets are added using AOMs to provide probe and reference combs for spectral normalization. The probe (after passing through the gas cell) and reference combs are combined and then mixed with the LO comb. Multiheterodyne signals are detected on a 700 MHz NIR detector and digitized at rates up to 3 GHz depending on the comb spectral bandwidth. For the remote sensing experiments, scatter from the Rocky Mountain Flatiron at a distance of ≈2.8 km is detected using a 28 cm telescope and photomultiplier tube for photon counting. In this case, the separate reference arm is not used. The signal and LO combs are combined and amplified to 13 mW before beam expansion and transmission to the atmosphere [8]. A small portion to generate the reference comb is fiber split after the final amplifier. In all cases, the data are streamed to memory with bandwidths up to 1.6 mega-sample/sec and processed and averaged in real time using a 56 hyper-threaded core computer before saving to disk.

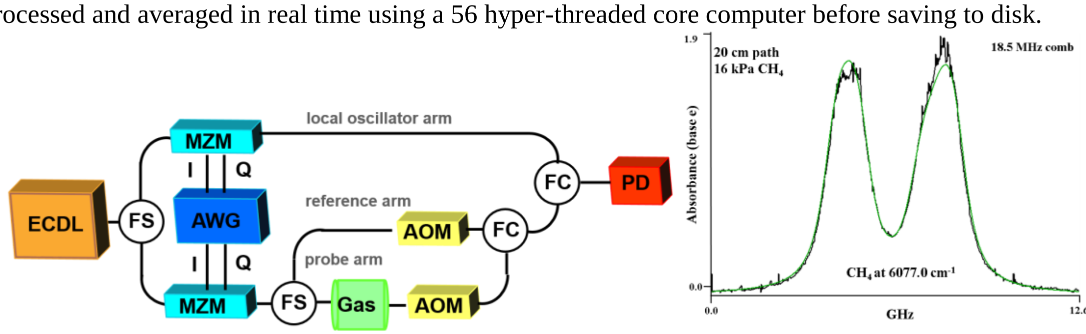

Fig. 1. Multiheterodyne spectrometer used for low pressure gas and remote sensing studies. ECDL, external cavity diode laser; FS, fiber splitter; FC: fiber coupler; MZM: dual parallel Mach Zehnder electro-optical phase modulator with in-phase (I) and quadrature (Q) inputs; AWG: arbitrary waveform generator; AOM: acousto-optical modulator; PD: photo-diode. Multiheterodyne spectrum of CH_4 (right) consisting of the average of 2 independent combs each normalized to its corresponding reference comb.

For the gas cell work, three frequency combs were produced using AM+PM waveforms. The waveform frequencies of first comb and second comb were 666.66667 MHz and 666.61667 MHz, respectively. The AM period of probe signal (third comb) is 54 ns to generate fine teeth with spacing 18.518 MHz and the period of the second comb (LO comb) is the same as the sampling period of 2.5 ms. The AM/PM combs were optimized (in an iterative way using genetic algorithms) by variation of the amplitudes, phases and pulse widths/shape of the fundamental and two harmonics over the sampling period. The digitized heterodyne waveform is Fourier transformed to give independent sets of rf combs for each of the probe and reference arms. A secondary set is also within the detection bandwidth but is reversed and offset relative to the first set. Each of the four combs contain nearly 700 teeth covering a 13.3 GHz region. For each set, the probe and reference combs are sorted, integrated and then normalized to the corresponding reference signals.

The amplitude transmission versus detuning of a multiheterodyne spectrum of CH_4 at 6077 cm^{-1} is shown in Fig. 1 (right). The spectrum was obtained using 20 cm long sample cell filled with 16 kPa of methane at room temperature and was acquired in 40 ms. The integrated intensity and doublet structure are in good agreement with predictions from the HITRAN database shown superimposed.

For the remote sensing study of CO_2 at 6241.40 cm^{-1}, the interoferograms of the probe (PMT) and reference signals are shown in Fig. 2 after time shifting the coadded probe interoferograms for the round trip transit time. The AOM frequency was 50 MHz to minimize the incoherence from the scattering. The MZM were driven at 1.000 GHz and 1.001 GHz giving an interferogram scan rate of 1 MHz to minimize effects of atmospheric turbulence. The data were acquired in <8 mins at an average count rate of $>10^6$ counts/sec. Phase jitter and drift in the system were corrected in real time using cross-correlations of the reference data with initial saved data. These same shifts were applied to the binning of the photon counts during accumulation. While the CO_2 fractional absorption is approximately correct, the large scatter that still remains is likely associated with reference channel normalization and wavelength decorrelation issues which are being addressed using a random distribution of relative phase shifts and stepped motion of the transceiver.

AWG-driven MZM generated combs enable agile changes in the comb spacing and bandwidth across a wide range (<1 MHz to >20 GHz) and therefore increase the utility of these methods for high resolution applications.

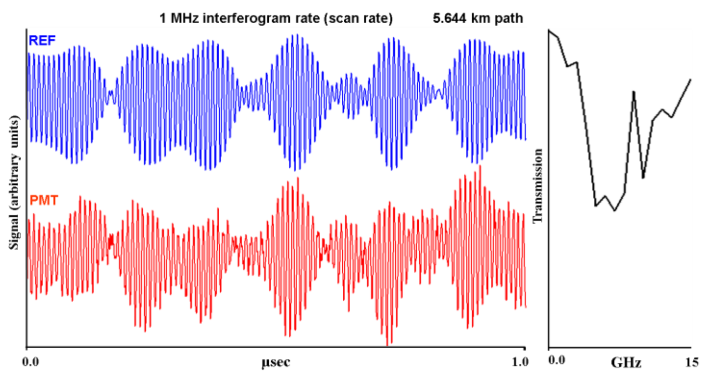

Fig. 2 10^8 averaged reference (top left) and coadded photon counting (bottom left) interferograms of CO_2/H_2O near 6241.4 cm^{-1} over 8 mins (20% throughput) acquired at 1 GHz sampling rate (left) and the normalized transmission spectrum (right) with fifteen 1 GHz comb teeth.

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