Electric Field Sampled Dual Frequency Comb Spectroscopy in Real Time Based on Ultrafast Cr:ZnS Laser Platform

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Abstract: We report the first demonstration of electro-optical sampling using two mutually coherent Cr:ZnS frequency combs. We recorded comb-mode resolved spectra of ammonia (8 – 12 μm) with high dynamic range and in real time. © 2022 The Author(s)

Electro-optic sampling (EOS) is a detection method that uses near-infrared (NIR) pulses to probe electromagnetic transients in the terahertz (THz) or mid-IR (MIR) domains [1, 2]. The nonlinear interaction results in a field–dependent change of the polarization state of the sampling NIR pulse that is resolved via ellipsometry using room temperature InGaAs photodetectors. Conventionally, EOS is used in combination with intra-pulse difference-frequency generation (IDFG) in quadratic nonlinear media that provides a robust source of ultrashort MIR/THz pulses with an intrinsically stable carrier envelope phase (CEP). The EOS features an outstanding dynamic range because it directly measures the electric field of MIR/THz pulses. An appealing extension of the EOS method is its combination with dual comb spectroscopy (DCS) technique providing high spectral resolution and high speed. A framework for performing the EOS-DCS combination was presented in [3]. The authors showed comb-mode resolved and broad bandwidth MIR spectroscopy using the averaging time of 88 min. Here we present a new platform for EOS-DCS that enables the comb-mode resolved measurements in real time.

Our approach is based on few-cycle Cr:ZnS laser oscillators at the MIR central wavelength λ = 2.4 μm [4]. The main advantages of this new laser platform are (i) access to ultra-broadband spectral coverage in the MIR (3–30 μm) that can be achieved with just two nonlinear crystals (ZGP and GaSe), (ii) high IDFG conversion efficiency reaching 10% in ZGP thanks to the favorable matching of group velocities between the 2.4-μm input and IDFG output, and (iii) the ease of producing 1-cycle sampling pulses at the convenient NIR wavelength 1.2 μm via second harmonic generation (SHG) and subsequent pulse compression.

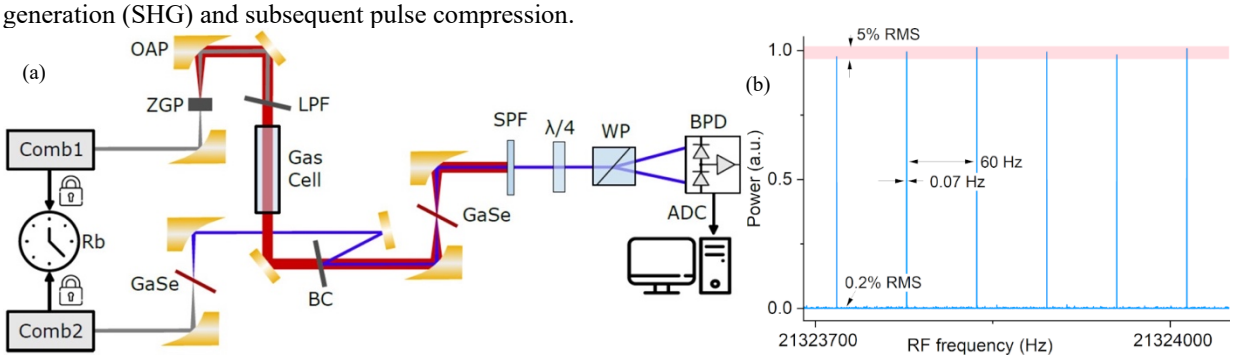

Fig. 1 (a) Schematic of the EOS-DCS MIR spectroscopy setup; SPF, short-pass filter; λ/4, waveplate; WP, Wollaston polarizer; OAP off-axis parabolas. (b) A detail of comb-mode resolved spectrum.

Our setup (Fig. 1a) includes a pair of 2-cycle Cr:ZnSe comb sources (central wavelength 2.4 μm, average power 3 W, pulse width <20 fs, pulse repetition frequency f_{rep} = 80 MHz). The high mutual coherence between two combs (effective coherence time >10 s) is achieved by their referencing to a common single frequency laser at the wavelength 1064 nm. The measured beatings between the combs and the optical reference as well as the combs' offset frequencies are phase-locked to a Rb clock. The pulse train from the first comb is coupled to an IDFG stage based on a 3-mm-long ZGP crystal. The IDFG output with 100-mW average power is long-pass filtered (LPF) to 8 – 12 μm and sent through a gas cell filled with 1% ammonia (NH_3) diluted in N_2 at a total pressure of 380 Torr. The beam from the second comb source is focused on a 30-μm thick GaSe crystal to produce 36 mW of second harmonic pulses (the obtained SHG spectrum supports 12-fs pulses however we did not do the pulse width metrology). The MIR and SHG beams were superimposed on a beam combiner (BC) and focused on a 340-μm thick GaSe crystal to produce an EOS signal, which is detected by a balanced InGaAs photodetector (BPD). The detected signal (EOS trace) is processed to obtain a broadband high-resolution spectrum modulated by molecular absorption in the gas sample.

To resolve the MIR comb-mode structure (Fig. 1b), we digitized streams of data with 16-bit resolution in a time window of 12 s at the sampling interval 12 ns. In total we acquired 700 traces spaced by 17 ms (corresponding to the repetition frequency offset Δf_{rep} = 60 Hz). The fast Fourier transform (FFT) of this signal, reveals – in power units –

the comb structure with the finesse of 750, the RMS fluctuation of the teeth power of 5%, and the RMS noise between the teeth of 0.2%.

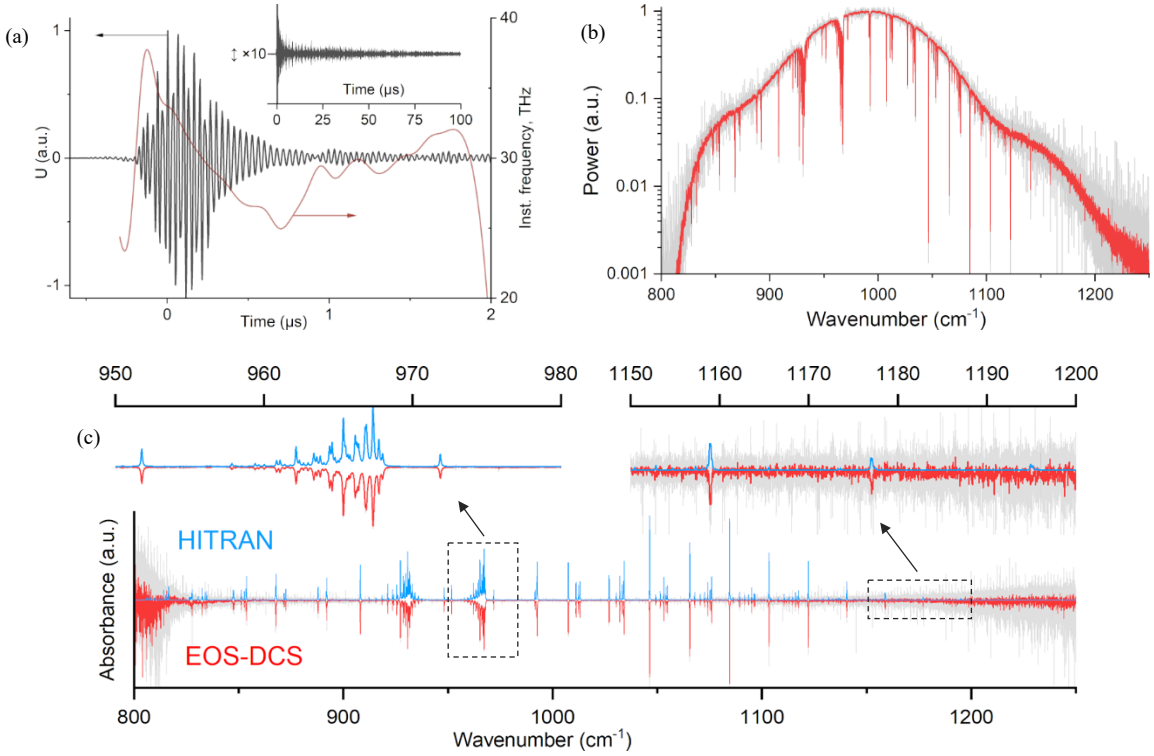

Fig. 2 (a) The central part of the averaged EOS trace and an estimated instantaneous optical frequency. The insert shows the trailing free induction decay of molecular absorbents. (b) MIR spectrum corresponding to the FFT of the measured waveform. (c) Absorbance spectrum obtained by subtracting the baseline, approximated with a polynomial fit. Gray background, no apodization (full resolution 0.0027 cm^{-1}); red line apodization of the time series to the resolution 0.027 cm^{-1}; blue lines HITRAN simulation (inverted for clarity). Inserts show details of the spectra near 10.4 μm (960 cm^{-1}) and 8.5 μm (1170 cm^{-1}).

To obtain a broadband molecular spectra we averaged EOS traces with an oscilloscope with 12 bit resolution. Figure 2a shows an average of 10240 traces. The obtained EOS trace indicates MIR pulses that are chirped by the ZGP crystal, Ge filters and gas cell's windows. Figure, 2b shows a raw spectrum obtained by the FFT of the acquired EOS trace. The spectrum spans 360 [425] cm^{-1} at 20 [30] dB with respect to the main peak that corresponds to 135,000 [159,000] comb modes. The derived absorbance spectrum of the gas sample is illustrated in Fig. 2c and compared with a simulated spectrum from the HITRAN2020 database. We observe a good fit to the HITRAN simulation of peak positions, linewidths, and relative peak intensities. Also, without apodization, we observe measurable spectroscopic signals at about -20 dB level that confirms the high dynamic range attainable with the EOS method.

In summary, this is the first, to the best of our knowledge, demonstration of IDFG/EOS dual comb spectroscopy based on ultrafast 2.4-μm laser platform. We recorded high-resolution (80 MHz, 0.0027 cm^{-1}) spectra of ammonia with the frequency scale referenced to a Rb clock. For the strongest peaks of ammonia (Fig. 2c), we obtained the signal-to-noise ratio of SNR=81 for just 12-sec averaging time and SNR>20 for 1-sec averaging, which opens up the possibility of real-time molecular detection with the full (80-MHz) resolution. These results set the stage for high-speed comb-mode resolved measurements in the MIR at room temperature.

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