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Direct Comb Multi-heterodyne Interference Spectroscopy

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ABSTRACT

We present a comb-based spectroscopic method that enables simultaneous detections of multiple gases by adopting an erbium-doped fiber femtosecond laser as a single broadband probing beam. The method takes multiple continuous-wave diode lasers as the frequency references, each being assigned to its distinct gas absorption line. The interference of the probing femtosecond laser with the diode lasers produces multi-heterodyne beats in the radio frequency domain, which are captured using a high-speed photodetector and electronically processed to identify the absorption lines of interest with a comb-mode spectral resolution. The experimental result of this study demonstrates that two gas absorption lines of H^{13}CN and $^{12}\text{CO}_2$, separated by a 23 nm spectral offset, can be detected concurrently at a 10 μs update rate with a 100 kHz spectral resolution. The proposed method finds applications in not only fundamental spectral line measurements for atomic and molecular physics but also diverse practical uses for remote sensing of trace and toxic gas molecules.

Keywords: optical frequency comb, high-resolution spectroscopy, direct comb spectroscopy.

1. INTRODUCTION

Laser absorption spectroscopy is a powerful tool for both fundamental spectroscopy, such as accurate measurement of two photon transition^{1,2} and remote sensing of trace gases as a coherent LIDAR³⁻¹¹. Furthermore, a variety of applications have emerged that require rapid sensing of target molecules including toxic gas monitoring, breath analysis^{4,11}, explosive detection⁵ and chemical reaction process monitoring. within the last decade, the invention of the stabilized optical frequency comb has revolutionized the field of spectroscopy and allowed measurements for fast, broadband and precise spectroscopy. Many methods have been developed to utilize the evenly spaced comb structure corresponding to millions of CW laser modes in broadband spectroscopy.

Here, we demonstrate CW laser referenced direct comb multi-heterodyne interference spectroscopy (DC-MIS) for rapid, multiple-molecular detection. This technique is capable of multispecies gas detection by measuring multiple heterodyne beats between a broadband frequency comb and multiple CW lasers with slightly different heterodyne frequencies for probing comb modes simultaneously. It employs just a femtosecond laser and CW lasers without any optical phase-locked loops, which is simple and versatile, and optical filtering of a probing optical frequency comb around the absorption lines of interest improves the sensitivity, thus signal-to-noise-ratio (SNR) of resolved comb spectrum and the efficiency related to the number of data points by removing unnecessary comb modes covering broad bandwidth. This technique is tested by measuring each absorption line profile of hydrogen cyanide (H^{13}CN) near 1550 nm and carbon dioxide ($^{12}\text{CO}_2$) near 1572 nm simultaneously by resolving comb modes of a femtosecond laser at measurement times of 10 μs , with the aim of rapid, multiple-molecular detection eventually.

2. OVERVIEW OF DC-MIS

A measurement principle of the DC-MIS is depicted in Fig. 1. An optical frequency comb which contains the information of interesting absorption lines is coupled with CW lasers. In our investigation, two distributed feedback (DFB) lasers which have different center wavelengths were utilized to measure two different kinds of gas molecules simultaneously. As shown in Fig. 1a, if we denote f_a as an optical frequency of the first DFB laser and f_b as that of the second DFB laser, each heterodyne frequency with the closest mode of the optical frequency comb can be defined as Δf_a and Δf_b respectively. Here, we initially set these beat frequencies to satisfy the condition of $\Delta f_a < \Delta f_b < f_r/2$ to determine the sign of each comb mode and separate beat signals generated from different absorption lines by monitoring the beat signals of RF spectrum. With a coupling between an optical frequency comb and DFB lasers, RF beat spectrum can be obtained by the Fourier-transform of the interferogram acquired from a high-speed photodetector and a high-speed oscilloscope. From our definition, beat frequencies between evenly spaced optical modes of a frequency comb and DFB lasers are denoted as $n \cdot f_r + \Delta f_a$ and $(n+1) \cdot f_r - \Delta f_a$ from the first DFB laser (red dashed line) and $n \cdot f_r + \Delta f_b$ and $(n+1) \cdot f_r - \Delta f_b$ from the second DFB laser (blue dashed line) as shown in Fig. 1b, where n is an integer. As absorption spectrum is represented in RF domain, its absorption line profiles of multi-heterodyne beats are folded in a positive side and by using overtones of a repetition rate as a reference frame, RF beat signals can be classified as lower or higher frequency components with respect to the optical frequency of a DFB laser. Here, lower frequency components are set to locate below the half of the repetition rate ($f_r/2$), which have $n \cdot f_r + \Delta f_a$ and $n \cdot f_r + \Delta f_b$. Higher frequency components above the half of the repetition rate, which have $(n+1) \cdot f_r - \Delta f_a$ and $(n+1) \cdot f_r - \Delta f_b$. Therefore, lower frequency components should be unfolded to the negative side in RF domain regarding optical frequencies of DFB lasers as zero to get absorption line profiles as shown in Fig. 1c. The final step is the determination of optical frequencies of DFB lasers; f_a and f_b to convert absorption spectra in RF domain into optical frequencies.

Figure 1. Measurement principle of DC-MIS. (a) Multi-heterodyne interference between an optical frequency comb and two DFB lasers adjacent to each absorption line of target molecules respectively. (b) RF beat spectrum based on multi-heterodyne down conversion between a probing optical frequency comb and reference two DFB lasers. (c) Data unfolding of RF beat spectrum to reconstruct absorption spectra of a probing optical frequency comb.

3. SYSTEM CONFIGURATION OF DC-MIS

For this study, we constructed a fiber spectrometer for DC-MIS to measure two kinds of gas molecules simultaneously. A femtosecond laser of which repetition rate is stabilized to a Rb clock with a value of 100 MHz is amplified and filtered by erbium doped fiber amplifier (EDFA) and two FBGs to increase the power per comb mode which makes this technique based on optical frequency comb versatile in many instances and consequently to enhance the SNR of beat signals under the detector's power limit. Two FBGs have center wavelengths of 1549.73 nm (FBG1) and 1572.01 nm (FBG2) at same filtering bandwidth of 25 GHz respectively. Filtered femtosecond lasers, which are used as a probe beam, pass through a multi-gas cell which consists of 10-torr H^{13}CN gas and 320-torr $^{12}\text{CO}_2$ gas. Here, target absorption lines are a P10 line of H^{13}CN gas molecule which has center wavelength of 1549.73051 nm and an absorption line of $^{12}\text{CO}_2$ gas molecules which has center wavelength of 1572.01797 nm. After passing through the multi-gas cell, a probing frequency comb is combined with two CW lasers for the purpose of generating multi-heterodyne interference signal. A center wavelength of each CW laser was set close to that of a target absorption line to measure full span of absorption lines within the bandwidth of a detector. In the experiment, two DFB lasers having the 100 kHz linewidth at a center wavelength of 1550.0 nm and the 1 MHz linewidth at a center wavelength of 1572.0 nm respectively were used as local oscillators. With the coupling of two DFB lasers and frequency comb modes, interferogram was obtained with 10 μs measurement time using a high-speed photodetector having 9.5 GHz bandwidth and a digital signal oscilloscope having 40 GHz sampling rate. The obtained interferogram was Fourier-transformed to RF beat spectrum which has 100

kHz spectral resolution at 8 GHz spectral bandwidth. This RF beat spectrum gives spectral information of absorption lines where the frequency of each beat signal shows a relative value to the center frequencies of DFB lasers. So, in order to convert obtained RF beat spectrum into optical frequency, center wavelengths of DFB lasers should be determined accurately. A multi-wavelength meter which has an accuracy of 12.5 MHz (0.1 pm) was utilized to monitor center wavelengths of DFB lasers simultaneously with the measurement.

Figure 2. Overall hardware configuration of DC-MIS; f_r : pulse repetition rate, C: fiber coupler, DFB LD: distributed feedback laser diode, PC: polarization controller, EDFA: erbium doped fiber amplifier, FBG: fiber Bragg grating.

4. EXPERIMENTAL RESULTS AND DISCUSSIONS

To evaluate the performance of our spectroscopy system, it has been used to measure the absorption lines of two independent gas molecules, 10-Torr H^{13}CN and 320-Torr $^{12}\text{CO}_2$ gas molecules within a multi-gas cell in one measure. The sampled double-sided interferogram was processed by apodization with a Hanning function. It was then Fourier-transformed to obtain RF beat spectrum as shown in Fig. 3a. Measurement time was 10 μs , corresponding to 100 kHz spectral resolution, and maximum measurement bandwidth was evaluated to 8 GHz, that is limited by maximum bandwidth of oscilloscope. The $\times 40$ magnified view (Fig. 3b) reveals that the beat signals by frequency comb and two DFB lasers' interference were located between self-heterodyne signals of frequency comb, corresponding to the harmonics of repetition rate (100 MHz). There are four peaks at 10 MHz, 90 MHz (red line) and 30 MHz, 70 MHz (blue line), which are the beat signals by frequency comb and DFB lasers' interference at 1550 nm and 1572 nm spectral region, including information of the H^{13}CN and $^{12}\text{CO}_2$ molecular absorption lines respectively. The SNR of the multi-heterodyne beats was 18 dB, and data unfolding of the RF beat spectrum reconstructed the optical spectral features of probing frequency comb as shown in Fig. 3c, which is normalized absorption spectrum of a H^{13}CN absorption line under 10-Torr pressure (red line) and a $^{12}\text{CO}_2$ line under 320-Torr pressure (blue line). Reference spectrum (black line) was measured in initial experiment without multi-gas cell to extract the transmittance of each absorption line profile and thus, remove most of the distortion that was imposed on the measurement by the electrical frequency response, yielding traces Fig. 3d. The achieved frequency resolution is estimated to be 100 kHz, which is sufficient to resolve frequency comb modes with 100 MHz comb spacing as shown in Fig. 3c. Each center wavelength of DFB lasers that was monitored using multi-wavelength meter in synchronization to the oscilloscope was substituted to zero frequency of Fig. 3c to transfer the RF beat spectrum into optical frequency domain. Fig. 3d shows the normalized transmittance spectra of

H^{13}CN P10 absorption line (red line) and $^{12}\text{CO}_2$ absorption line (blue line) measured simultaneously. The spectrum shown in Fig. 3d clearly reveals the absorption lines of the H^{13}CN and $^{12}\text{CO}_2$ gas molecules at a resolution of 100 MHz (0.0008 nm) per datapoint corresponding to the repetition frequency of a probing frequency comb over a wavelength range of 22 nm, despite the detection bandwidth of 16 GHz (0.132 nm) for each absorption line. A fit based on a complex Voigt profile is also superimposed with the data points.

In conclusion, Our CW laser referenced direct comb multi-heterodyne interference spectroscopy (DC-MIS) demonstrates comb-resolved, rapid spectroscopy which offers 100 kHz resolution with 10 μs measurement time and measures multiple gas molecules in 22 nm away simultaneously. SNR is 18 dB for a measurement and It is anticipated that the DC-MIS can be exploited for rapid gas sensing application particularly toxic gas monitoring and chemical reaction process monitoring in addition to high-resolution spectroscopy for multispecies gas detection.

Figure 3. (a) Measured RF spectrum by the Fourier-transform of multi-heterodyne interference signals between a frequency comb and two DFB lasers. (b) Magnified views ($\times 40$) showing heterodyne beats with different intervals of 10 MHz in 1550 nm (red line), and 30 MHz in 1572 nm (blue line) between a comb and two DFB lasers. (c) Normalized absorption spectrum of a H^{13}CN absorption line of under 10-Torr pressure (red line) and a $^{12}\text{CO}_2$ line under 320-Torr pressure (blue line). Reference spectrum (black line) is measured in initial experiment to extract an absorption line profile and remove most of the distortion that was imposed on the measurement by the electrical frequency response. (d) Transmittance spectra of a P10 absorption line of H^{13}CN gas cell of 10-Torr pressure (red line) and an absorption line of $^{12}\text{CO}_2$ cell of 320-Torr pressure (blue line) over 23 nm spectral bandwidth.

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