Opto-Electronics Review 26 (2018) 103–107

Contents lists available at ScienceDirect

Opto-Electronics Review

journal homepage: http://www.journals.elsevier.com/opto-electronics review

Frequency-multiplexed gas sensing using chirped laser molecular spectroscopy

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ARTICLE INFO

Article history:
Received 1 December 2017
Received in revised form 20 February 2018
Accepted 23 February 2018

Keywords:
Laser spectroscopy
Heterodyne detection
Optical sensing
Gas sensing

ABSTRACT

A method for frequency-multiplexed multi-sample gas sensing is presented. It enables measuring multiple samples placed simultaneously in the setup, without any optical or mechanical switching. Samples are measured using heterodyne detection and signal from each sensing path is encoded at different carrier frequency. Subsequently, a signal from particular sample is retrieved through heterodyne beatnote demodulation at unique frequency. This technique is particularly suitable for real-time calibration of the sensor through a sequential (or simultaneous) detection of three signals: from unknown sample, reference sample and baseline. Basic setup is demonstrated and proof-of-concept experiments are presented. Very good agreement with spectra measured using standard tunable diode absorption spectroscopy is obtained.

1. Introduction

Laser molecular spectroscopy is a powerful tool in chemical analysis with various applications, including fundamental studies [1,2], environmental monitoring in point [3–6] and path-integrated [7,8] arrangements, isotope ratio measurements [9–11], breath analysis [12–15], safety [16–18], or industrial process control [19–22]. Using standard absorption-based methods such as direct tunable diode laser absorption spectroscopy (TDLAS) [23–26] or wavelength modulation spectroscopy (WMS) [27,28] high sensitivity/precision can be obtained. However, in many applications a more critical issue is the long-term stability of the system. Opto-mechanical alignment changes, beam pointing instabilities or electrical drifts affect accuracy of the measurement and they usually cannot be removed through signal averaging. In typical spectroscopic instrumentation this issue is addressed by periodic calibration of the system. This can be realized, e.g., using two separate measuring paths, one for unknown sample and another for calibration gas. Alternatively, the sample and calibration gas can be measured in turns, using only one sensing path. Both approaches have advantages and drawbacks. Two separate paths require two detectors which can be expensive, especially when sensing is performed in the mid-infrared spectral region. Detectors may have different responsivities, offsets and drifts. On the other hand, single path approach cannot be directly implemented, e.g., in open-path sensing. Up to date several alternative methods for sensor calibration has been proposed. In Ref. 29 a carbon monoxide sensor was presented in which photodetector housing was additionally filled with methane as a reference gas. Methane transition near 2.3 μm was used for a wavelength scale calibration and for monitoring how laser parameters drift in time. Similar concept was also presented in Refs. 30 and 31 where an additional reference gas cell was placed in-line with an open-path multi-pass cell to enable continuous, real-time calibration of the WMS-based measurement. In Ref. 30 a cell was filled with ethylene that has absorption line in the vicinity of a target transition of ammonia near 9.06 μm. In Ref. 31 a cell with acetylene was used to calibrate open-path measurement of nitrous oxide and carbon monoxide near 4.5 μm. In both cases multiple harmonic WMS signals were used to separate spectral features of target species from signals from reference gases. In Ref. 32 a revolving cell was used to measure unknown sample, calibration gas and zero-gas. This enabled a real-time calibration of a WMS-based measurement of carbon dioxide near 4.23 μm.

Another group are techniques which enable simultaneous detection of multiple samples through multiplexing of spectroscopic signals in frequency or time domain. In Refs. 33–35 frequency multiplexing of sensors placed in a ladder topology was demonstrated. With an external triangular intensity modulation of light from a tunable laser and with additional passive delay lines with each gas sample, useful signals could be encoded into different frequencies at the photodetector's output. These sig-

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https://doi.org/10.1016/j.opelre.2018.02.004
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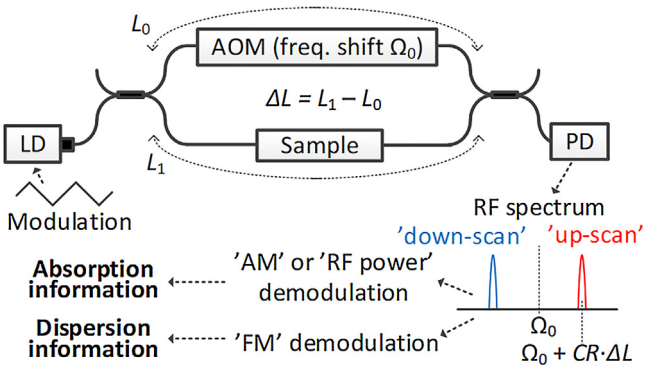

Fig. 1. Basic setup for chirped laser spectroscopy as demonstrated by Wysocki and Weidman in Ref. 38 ('single frequency beam configuration'). LD – laser diode, PD – photodiode, *CR* – chirp rate, AOM – acousto-optical modulator/frequency shifter.

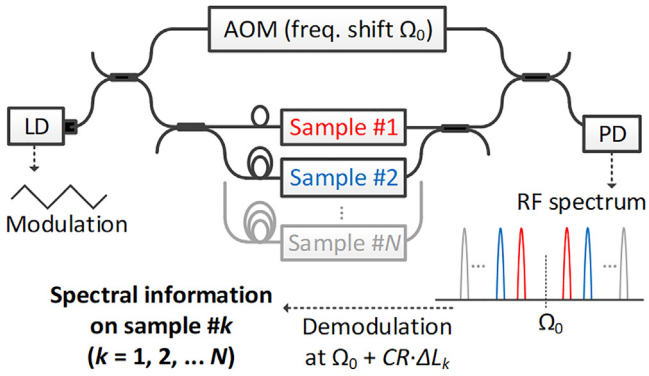

Fig. 2. Schematic diagram of a setup for frequency-multiplexed chirped laser spectroscopy. Due to (relatively small) differences in lengths of delay lines spectroscopic information from each sample is encoded at different carrier frequency Ω_k. Information on particular sample can be retrieved through demodulation at specific frequency, without the need of any optical or mechanical switching within the setup.

nals were subsequently retrieved using an electrical mixer and a band-pass filter. Similar approach was demonstrated in Ref. 36 using a chirped laser dispersion spectroscopy (CLaDS). Unfortunately, in all four examples relatively long delay lines must be used, thus a straightforward transfer of this idea into a mid-infrared spectral region would be challenging. Another interesting idea for simultaneous analyzing of two samples was recently demonstrated in Ref. 37. Differential Optical Dispersion Spectroscopy (DODiS) enables simultaneous detection of molecular absorption and dispersion in the setup. Furthermore, when a proper configuration is used, a detected absorption signal corresponds to the cumulative absorption from two analyzed samples, and the measured dispersion signal corresponds to the differential dispersion. These can be used to retrieve the sum and the difference of concentrations, consequently providing all information about both individual concentrations of gas molecules in the samples. DODiS has potential to become a useful tool in applications where exactly two samples have to be measured and cross-compared, ideally with a single measurement process (e.g., isotopic ratiometery). Unfortunately, using it in setups with more than two samples requires additional acousto-optical modulators, increasing costs and complexity.

In this paper we present a frequency-multiplexed multi-sample gas detection approach that grows upon CLaDS and DODiS techniques. It enables measuring multiple samples placed in a ladder topology (similar to [33–35]) using only relatively short delay lines (in the orders of meters). Thus, it can be implemented in both near- and mid-infrared spectral regions. It may be easily scaled up (increasing number of sensing paths does not require any additional active optical components) and enables detection of both molecular absorption and dispersion. In this work a basic setup is demonstrated and proof-of-concept experiments are presented. Switching between three independent measurements is performed without any opto-mechanical changes in the setup, it only requires changing demodulation frequency. Very good agreement with spectra measured using standard tunable diode absorption spectroscopy is obtained.

2. Chirped laser spectroscopy: principle

A basic setup for chirped laser spectroscopy as demonstrated by Wysocki and Weidman [38] is presented in Fig. 1. It consists of a single-frequency laser source (typically a DFB laser diode), and a Mach-Zehnder interferometer with an acousto-optical modulator/frequency shifter (AOM) in one arm and a gas sample in the other. Output of the interferometer is focused onto a fast photodiode that records a heterodyne beatnote. As the laser is frequency scanned (chirped) across target transition spectroscopic information about molecular absorption and dispersion is encoded into amplitude and frequency of the beatnote. It can be retrieved through AM and FM demodulation (respectively) at $\Omega = |\Omega_0 + \Delta L \cdot CR|$, where Ω_0 is AOM's frequency, ΔL is the unbalance of the interferometer, and *CR* is the chirp rate.

3. Frequency-multiplexed spectroscopy of multiple sampleS

An optical arrangement for a frequency-multiplexed chirped laser spectroscopy is shown in Fig. 2. It is a modified version of the setup shown previously. Multiple samples can be placed in it using fiber-based couplers/splitters. Optical path for each sample has to have unique length (short delay lines can be used if necessary). As a result, the signal from a sample k ($k = 1, 2, \ldots N$, where N is the number of samples) is encoded into RF frequency that can be expressed as $\Omega_k = |\Omega_0 + \Delta L_k \cdot CR|$. Consequently, all N samples are probed simultaneously within single laser scan, producing N signals which can be conveniently separated in the RF domain. Detecting signal from particular sample can be accomplished by selecting an appropriate demodulation frequency, in a similar way as stations are selected in AM or FM radio.

3.1. Experimental setup

For experimental demonstration of frequency-multiplexed multi-sample spectroscopy an optical setup with three sensing arms was built. A DFB laser diode operating near 1651 nm (NTT Electronics, model NLK1U5EAAA; maximum injection current ≈120 mA) was used to target R4 transition in the $2\nu_3$ band of methane. It was driven and temperature-stabilized at ~20 °C using commercial laser diode controller (Arroyo Instruments, model 6305). No sample was placed in arm #1, therefore it enabled recording a baseline signal (a 'zero gas' measurement). In arms #2 and #3 two samples were placed, both were 25 mm long cells with methane balanced with nitrogen at 740 Torr, with concentrations of 20% and 4%, respectively. Gas cells were manufactured by Wavelength References. Their windows were tilted and had anti-reflective coating to prevent reflections that can lead to unwanted fringes. Setup was built using several standard fiber couplers/splitters which were chosen so that heterodyne signals from all three arms had similar amplitudes (between −25 dBm and −35 dBm). Short pieces of a standard single-mode fiber SMF-28 were used to guarantee that the lengths of all arms are different ($\Delta L_1 \approx 1.5$ m, $\Delta L_2 \approx 6.4$ m, $\Delta L_3 \approx 11.7$ m). A fiber-coupled acousto-optical modulator (AA Opto-electronic, model MA40-IIR120) was used to provides frequency shift of $\Omega_0 = 40$ MHz. It was driven with