A compact mid-infrared dual-gas CH$_4$/C$_2$H$_6$ sensor using a single interband cascade laser and custom electronics

Weilin Ye $^{a,b}$, Chuantao Zheng$^{a,c,*}$, Frank K. Tittel$^a$, Nancy P. Sanchez$^d$, Aleksander K. Gluszek$^{a,c}$, Arkadiusz J. Hudzikowski$^{a,e}$, Minhan Lou$^a$, Lei Dong$^f$, Robert J. Griffin$^d$

$^a$ Department of Electrical and Computer Engineering, Rice University, 6100 Main Street, Houston, TX 77005, USA, $^b$ College of Engineering, Shantou University, 254 Daxue Road, Shantou 515063, P. R. China, $^c$ State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, 2699 Qianjin Street, Changchun 130012, P.R.China, $^d$ Department of Civil and Environmental Engineering, Rice University, 6100 Main Street, Houston, TX 77005, USA, $^e$ Laser & Fiber Electronics Group, Faculty of Electronics, Wroclaw University of Technology, Wybrzeze Wyspianskiego 27, 50-370 Wroclaw, Poland, $^f$State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, Taiyuan 030006, P.R.China

*Corresponding author. zhengchuantao@jlu.edu.cn

ABSTRACT

A compact mid-infrared (MIR) dual-gas sensor system was demonstrated for simultaneous detection of methane (CH$_4$) and ethane (C$_2$H$_6$) using a single continuous-wave (CW) interband cascade laser (ICL) based on tunable laser absorption spectroscopy (TDLAS) and wavelength modulation spectroscopy (WMS). Ultracompact custom electronics were developed, including a laser current driver, a temperature controller and a lock-in amplifier. These custom electronics reduce the size and weight of the sensor system as compared with a previous version based on commercial electronics. A multipass gas cell with an effective optical length of 54.6 m was employed to enhance the absorption signal. A 3337 nm ICL was capable of targeting a C$_2$H$_6$ absorption line at 2996.88 cm$^{-1}$ and a CH$_4$ line at 2999.06 cm$^{-1}$. Dual-gas detection was realized by scanning both the CH$_4$ and C$_2$H$_6$ absorption lines. Based on an Allan deviation analysis, the 1σ minimum detection limit (MDL) was 17.4 ppbv for CH$_4$ and 2.4 ppbv for C$_2$H$_6$ with an integration time of 4.3 s. TDLAS based sensor measurements for both indoor and outdoor mixing ratios of CH$_4$ and C$_2$H$_6$ were conducted. The reported single ICL based dual-gas sensor system has the advantages of reduced size and cost without influencing the mid-infrared sensor detection sensitivity, selectivity and reliability.

Keywords: Mid-infrared, Interband cascade laser, Dual-gas detection, Wavelength modulation

1. INTRODUCTION

Methane (CH$_4$) is a main contributor to the greenhouse effect and a safety hazard in the production of chemicals. Hence the monitoring of CH$_4$ concentration levels is critical in urban or rural areas [1, 2]. Ethane (C$_2$H$_6$) [3, 4] is the second-largest component of natural gas after CH$_4$ and is mainly used in the chemical industry. Simultaneous detection of C$_2$H$_6$ and CH$_4$ is an effective method to discriminate the CH$_4$ origin between thermogenic (e.g. natural gas production) and biogenic sources (e.g. landfills, wetlands). Therefore, a dual-gas sensor was developed to perform the detection of CH$_4$ at an atmospheric concentration of ~ two parts-per-million by volume (ppmv) and C$_2$H$_6$ of several parts-per-billion by volume (ppbv) to tens of ppbv.

Infrared laser spectroscopy [5-9] is advantageous compared to other detection methods in terms of cost, size and no sample pretreatment. Furthermore, TDLAS can achieve high-precision sensing capabilities and fast response. TDLAS [10-12] enables direct concentration measurements and has proven to be an excellent tool for trace gas detection in...
various applications. TDLAS requires a tunable laser with narrow linewidth to reach a high detection sensitivity in the infrared spectral range. Quantum cascade lasers (QCLs) [13] and interband cascade lasers (ICLs) [14] which can provide continuous-wave (CW) output power levels in the mW range and above are the optimum choice for TDLAS.

In previous studies, an ICL emitting at 3291 nm was used in a CH₄ detection system [15] and an ICL emitting at 3337 nm was used in a C₂H₆ detection system [16] for monitoring CH₄ mixing ratios at natural gas vehicle fueling stations. However, in order to detect both CH₄ and C₂H₆ simultaneously, the two systems with two data acquisition (DAQ) cards, vacuum pumps, pressure controllers, commercial lock-in amplifiers, commercial QCL and ICL current drivers and temperature controllers required high electrical power consumption and large sensor footprint. Therefore, a compact dual-gas CH₄/C₂H₆ sensor system with small sized custom electronics was developed. A single ICL emitting at 3337 nm was selected and modulated to target CH₄ and C₂H₆ absorption lines at 3334 nm and 3336 nm, respectively. Furthermore, a LabVIEW program controlled signal generator, signal acquisition and lock-in amplifier, a compact digital current driver and temperature controller were developed to reduce the system size and electrical power-consumption.

2. SENSOR STRUCTURE AND DESIGN DETAILS

2.1 Absorption line selection

Both CH₄ and C₂H₆ have fundamental vibrational bands at 3.34 μm. Absorption spectra of 10 ppbv C₂H₆, 2 ppmv CH₄, and 2% H₂O at 100 Torr gas pressure and a 5460 cm optical path length, based on the 2012 HITRAN database are depicted in Fig. 1(a). An optimal diagnostic window at 2996.88 cm⁻¹ was identified for C₂H₆ detection. There are two strong CH₄ absorption lines located at 2998.99 cm⁻¹ and 2999.06 cm⁻¹. However, two H₂O lines in the neighborhood of these absorption lines can be problematic. The 2998.97 cm⁻¹ H₂O line almost overlaps with the 2998.99 cm⁻¹ CH₄ line, and therefore the 2999.06 cm⁻¹ CH₄ absorption line was selected for optimum detection of this trace gas.

The ICL was operated at 10 °C employing a thermal electrically cooler (TEC). A high precision wavelength meter (Bristol 627) was employed to measure the output wavenumber which is shown in Figure 1(b). The ICL drive current was between 32 and 47 mA in order to cover both CH₄ and C₂H₆ absorption lines.

![Fig. 1. (a) HITRAN based absorption spectra of C₂H₆ (10 ppbv), CH₄ (2 ppmv) and H₂O (2%) in a spectral range from 2996 cm⁻¹ to 3002 cm⁻¹ at a pressure of 100 Torr and an absorption path length of 54.6 m. C₂H₆, CH₄, and H₂O absorption lines are shown in green, blue and red, respectively. (b) Plot of the ICL emission wavenumber as a function of the ICL drive current at 10 °C.](image-url)

2.2 Sensor System

The dual-gas CH₄/C₂H₆ sensor architecture is depicted in Fig. 2. The sensor unit includes an optical and an electrical component. In the optical part, an alignment laser was employed as a guide beam in order to facilitate the optical alignment process. The infrared beam from the ICL and the visible beam from an alignment laser (AL) were combined by a dichroic mirror (DM). A mode matching lens (L) and two adjustable plane mirrors (M1 and M2) were used to
couple and focus the combined beam into the 54.6 m multi-pass gas cell (MPGC). After 432 reflections, the output beam was focused onto a TEC mercury-cadmium-telluride (MCT) photodetector by a parabolic mirror.

In the electrical sensor sub-system, a WMS technique was used for the detection of both trace gases, which required a scan and a modulation signal to drive the ICL. These two signals were generated and added by a LabVIEW program installed in a laptop and the signal output was processed by a National Instruments DAQ card applied to a digital ICL current driver. The temperature and current of the ICL were controlled by a custom temperature controller and current driver, respectively (see Sec. 2.4). Furthermore, the output signal from MCT detector was applied to the DAQ card and processed by the LabVIEW program. A LabVIEW based lock-in amplifier was developed to obtain the harmonic signals.

In addition, an oil-free vacuum pump was used to introduce the target gases into the MPGC and a pressure controller (PC) was used to control the pressure inside the MPGC. A water trap was used at the gas inlet, in order to avoid the interference of water vapor present in the sampled air.

![Fig. 2. Schematic of the dual-gas CH₄/C₂H₆ sensor based on a single CW, TEC ICL. ICL: interband cascade laser; DM: dichroic mirror; AL: alignment diode laser; M: plane mirror; PM: parabolic mirror; MCT: mercury-cadmium-telluride detector. PC: pressure controller; SA: signal acquisition; HE: harmonic extraction; SG: signal generation.](image)

**2.3 LabVIEW-based data-processing system**

A LabVIEW program was developed, whose function diagram is shown in Fig. 3. A modulation and a scan signal array were generated and superimposed as the laser driver signal to the digital-to-analog converter (DAC) module. The output signal from MCT detector was acquired by an analog-to-digital converter (ADC). This signal multiplied by two frequency-doubled orthogonal signals which were synchronized by the modulation signal. With low-pass filtering (LPF), biased adding operation (Bias) and square operation (Square), two orthogonal components were obtained. Then, using a second adding operation between the two components and a square root (Sqrt) operation, the R signal was obtained. For convenient data processing, the baseline from the R signal was removed via a subtraction operation, and the 2f signal was extracted to determine the gas concentration.

![Fig. 3. Function diagram of the LabVIEW-based laptop platform, which performs signal generation, signal acquisition and harmonic extraction.](image)
2.4 Customer electronics design

Custom board-level electronics (see Fig. 4(a)) with a size of 4.2 × 4.8 × 2.0 cm³ and a temperature controller (see Fig. 4(b)) with a size of 4.5 × 3.6 × 2.0 cm³ were developed. The output range of current driver is 0 – 300 mA and the ratio between input voltage and output current was adjusted to 20 mA/V. An accuracy of 0.001 °C can be obtained by means of the custom based digital temperature controller.

Fig. 4 Rice University customs electronics design. (a) Laser current driver with dimensions of 4.2 × 4.8 × 2.0 cm³ cm and a current range of 0 - 300 mA, (b) Temperature controller with dimensions of 4.5 × 3.6 × 2.0 cm³ and a short term stability ~ ± 0.001 °C.

3. PERFORMANCE OF SENSOR SYSTEM

In order to cover the two target absorption lines of CH₄ and C₂H₆ located at 2999.06 cm⁻¹ and 2996.88 cm⁻¹, respectively, a drive signal biased at ~ 2.0 V with a current range of 31–49 mA was applied to the ICL operating at 10 °C. This signal contained three parts. The first part and the third part were two ramp signals superimposed by modulation signals, which were used to scan the selected CH₄ and C₂H₆ line, respectively. The center part was an exponential signal used to connect the first part and the third part with a duration time of 1 s. Hence, the period of such a drive signal was ~ 4.3 s. A pressure of 100 Torr inside the MPGC was selected for optimum sensor operation. The direct output signal from the detector and the demodulated 2f signal from the lock-in amplifier with a time constant of ~ 16 ms for the 2.1 ppmv CH₄ and 0 ppbv C₂H₆ are shown in Fig. 5(a). The two signals for 0 ppmv CH₄ and 90 ppbv C₂H₆ are shown in Fig. 5(b). The two absorption peaks of CH₄ and C₂H₆ are included in both the absorption and the 2f signals with a single laser scan.

Fig. 5. Direct output signal from the MCT detector and demodulated 2f signal by the LabVIEW-based lock-in amplifier with a time constant of ~ 16 ms, (a) for a CH₄/C₂H₆:N₂ mixture with a CH₄ concentration of 2.1 ppmv and C₂H₆ concentration of 0 ppbv, and (b) for a CH₄/C₂H₆:N₂ mixture with a CH₄ concentration of 0 ppmv and a C₂H₆ concentration of 90 ppbv.

The dual-gas sensor was calibrated using 2.1 ppmv CH₄ and 1.14 ppmv C₂H₆ standard concentration cylinders mixed with pure nitrogen using a dilution system (Environsics S-4040). The concentration levels of four CH₄ samples (0, 300,
600, 900 ppbv) and four C2H6 samples (0, 30, 60, 90 ppbv) were measured and their results are shown in figures 6(a) and 6(b). For an observation period of < 10 min, the dual-gas sensor was confirmed to have a high accuracy with a small variation range for each measured concentration. Long-term measurements for 0 ppmv CH4 and 0 ppbv C2H6 were conducted for ~ 40 min. Allan deviations were obtained and shown as insets of figures 6(a) and 6(b), respectively. The Allan deviations for the two gases were 17.4 ppbv for CH4 and 2.4 ppbv for C2H6 for an averaging time of 4.3 s.

![Fig. 6](image)

**Fig. 6.** Measurement results of concentration levels of (a) four CH4 samples (0, 300, 600, 900 ppbv) and (b) four C2H6 samples (0, 30, 60, 90 ppbv). The insets in Fig. 6(a) and 6(b) exhibit the Allan deviation plots obtained from long-term measurements of 0 ppmv CH4 and 0 ppbv C2H6 samples for ~ 40 min, respectively, using the calibrated dual-gas sensor system.

## 4. ATMOSPHERIC CH4/C2H6 DETECTION

The dual-gas CH4/C2H6 sensor along with a meteorological station (Airmar 150WX) were deployed in a vehicle and employed for extensive monitoring of CH4/C2H6 atmospheric concentration levels in the Greater Houston area (GHA) during summer 2016. Fig. 7 (a) shows different areas in the GHA that were selected for investigating the potential occurrence of CH4 leaks related with the natural gas (NG) distribution system in the Houston area. These locations were classified as zones with high (H1 and H2), medium (MA1 and MA2) and low (L1 and L2) probability of occurrence of CH4 leaks. Fig. 7 (b) depicts the dual-gas sensor deployed in a vehicle during the field tests.

![Fig. 7](image)

**Fig. 7 (a)** Different selected areas in the GHA for investigating the occurrence of CH4 leaks. **(b)** Deployment of the CH4/C2H6 sensor system for mobile-mode monitoring of CH4 and C2H6 concentrations at selected GHA zones.

Fig. 8 (a) and (b) depict the spatial variation of the CH4 and C2H6 mixing ratios measured at one of the selected locations classified as with high (H1) probability of CH4 leaks on August 8, 2016. As can be seen, CH4 concentrations ranged
between 2.1 and 2.7 ppm while C$_2$H$_6$ levels were ~ 10 ppb with some observed spikes reaching up to 20 ppb. These observed increases in the C$_2$H$_6$ mixing ratios corresponded to instantaneous peaks likely caused by vibrations due to uneven road conditions.

Fig. 8 Field test with the reported dual-gas CH$_4$/C$_2$H$_6$ sensor system at a residential zone in central Houston (H1) on Aug. 8, 2016 measured (a) CH$_4$ and (b) C$_2$H$_6$.

5. CONCLUSIONS

A sensitive and selective sensor system based on a single CW DFB ICL was demonstrated for simultaneous detection of CH$_4$ and C$_2$H$_6$. An ICL with a wavelength of ~ 3.337 µm was employed to target two absorption lines of CH$_4$ and C$_2$H$_6$ within a narrow spectral range of ~ 3 cm$^{-1}$. The sensor system was first evaluated for individual CH$_4$ and C$_2$H$_6$ detection, with the ICL wavelength tuned to each gas absorption line center. Custom electronics were developed, including an ultra-compact laser current driver, a temperature controller and a lock-in amplifier which reduced the size and weight of the sensor system. An Allan deviation analysis yielded detection sensitivities of 17.4 ppbv for CH$_4$ and 2.4 ppbv for C$_2$H$_6$ for an integration time of 4.3 s due to the increased drift in both ICL temperature and power caused by the longer scanning time periods. Measurement results for outdoor atmospheric concentration changes of CH$_4$ and C$_2$H$_6$ were also reported. The demonstrated dual-gas sensor architecture shows the merits of simultaneous CH$_4$ and C$_2$H$_6$ detection with a single sensor system of significantly reduced size and cost without influencing the mid-infrared sensor detection sensitivity, selectivity and reliability.

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