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Multi-Species Continuous Gas Detection with Supercontinuum Laser at Telecommunication Wavelength

Abubakar I. Adamu, Manoj K. Dasa, Ole Bang and Christos Markos

Abstract— Supercontinuum (SC) lasers provide an excellent opportunity for absorption spectroscopy by virtue of their high brightness and large bandwidth. Here we present the detection of multiple industrial toxic gases from 1480 nm to 1700 nm, with a simple custom SC laser source. Using readily available optical components, we demonstrate an all-fiber system for the detection of ammonia (NH₃) and methane (CH₄) in a 2.4 m long hollow-core photonic bandgap fiber with 20-µm core diameter. The responsivity, selectivity, and performance of the system for continuous detection are demonstrated.

Index Terms— Optoelectronic and photonic sensors, Gas sensors, Environmental monitoring, Optical fiber sensors.

1. INTRODUCTION

Supercontinuum (SC) lasers are spatially coherent so-called white light lasers with an enormous bandwidth covering several octaves and an extreme spectral brightness, which is orders of magnitude higher than even synchrotrons [1]. The unique properties of the SC laser has allowed it to make a remarkable impact in the fields of applied spectroscopy [2], food control [3], biomedical sensing applications [4], [5], photo-acoustic and optical coherence tomography imaging [6]–[9], telecommunication [10], inspection of optical components and semiconductor chips [11]–[13], to name a few. The majority of the laser sources used in the above-mentioned reports are fiber-based, which makes them compact and robust and therefore ideal for also operation in harsh environments, such as in stand-off detections of explosives and chemicals [14], [15].

Gas sensors based on optical methods are remarkable for their longevity, high sensitivity, selectivity, and immunity to catalyst poisoning and environmental changes [16]. Additionally, their very short response time enables on-line real-time gas detection. One of the optical sensing techniques is spectroscopy, such analysis primarily involves techniques which rely on absorption and emission spectrometry [16]. Absorption spectroscopy relies on the Beer-Lambert law, which provides the relation for concentration-dependent molar absorptivity of photons at specific wavelengths [17], which is distinct for every gas because of their absorption fingerprints, thus enabling for high selectivity. This selectivity, coupled with the broadband spectrum of the SC source, allows for multi-species gas detection [18], [19]. Comparatively, absorption-based optical gas sensors have advantage over electrochemical based gas sensors or metal oxide-based sensors [20], [21] which typically target only a specific gas. Most cost-effective and commonly used gas sensors today are metal oxide sensors [16], whose device response depends on the redox reaction between the target gas and the oxide surface [16], [22]. This practically means that they suffer from a lack of selectivity [23] and short lifetime as a result of deteriorating metal oxide surface. Although such sensors can be very sensitive, their high sensitivity comes at a cost, because they often require high operation temperatures of up to typically a few hundred degrees Celsius [24], [25]. Furthermore, there is a constraint on other NH₃ sensors as to the maximum detection limit, with photoionization detectors having a response to 1000 ppm [26] and electrochemical based NH₃ sensors saturating at only 100 ppm [26], [27].

Unlike the aforementioned chemical reaction-based technique, spectroscopy-based sensors target the specific absorption of molecules, and because every gas has its distinct and unique absorption spectrum, the sensor can be highly selective. Thus, rather than cross-examining the response from arrays of sensors, one can deploy a single broadband SC source and target multiple gases. Other results have demonstrated the use of hollow core fiber for gas sensing, such as photothermal gas sensors [28] and very recently, a...
study from the same group on mode-phase-difference based photothermal sensing [29] demonstrated acetylene detection with non-commercial fibers. Although remarkable performance has been reported in the literature, there is no report on multiple continuous gas detection with single laser source. Our cheap and easy-to-build SC source provides a suitable solution for such applications since tunable quantum cascade lasers are not available in the near-IR.

Here we demonstrate a home-built SC laser suitable for the simultaneous detection of ammonia (NH$_3$) and methane (CH$_4$). These two gases are chosen because they are the most predominant gasses emitted in places of intense agriculture, such as dairy barns [30], livestock farming [31] and fertilizer industry [23]. Exposure to NH$_3$ can lead to many medical complications ranging from severe nasal irritation to death, depending on the concentration and the exposure time [23]. Farming being the major contributor of NH$_3$ in the atmosphere accounts for over 50% of global NH$_3$ emission [31], [32]. Furthermore, about 50% of this NH$_3$ emission ends up as a form of environmental pollutant. Agriculture is also the second highest contributor to CH$_4$ emission, accounting for up to 31% of total emission in the US for example [33]. The emission of CH$_4$ is considered among the primary contributors to Earth warming [34]. Therefore, having a gas sensor that can continuously detect and monitor the emissions of these gases is of great importance. We present a cost-effective method for detecting these two gases using optical-based absorption spectroscopy in a hollow-core photonic band gap (HCPBG) fiber. For any optical fiber sensor to be practical and deployable in the field, it is necessary for it to be compact and all-fiberized. To this end, we demonstrate a complete all-fiber system based on commercial telecom range components, where gas entry and exit into the 20 μm hollow core is achieved with a mechanical splice, between the single-mode fiber (SMF) (SMF-28, Corning) and HCPBG. One advantage of operating in the telecommunication wavelength regime is the availability of high-quality components at low-cost, thanks to the advanced level of development in optical telecommunication technology. Therefore, the overall developed system costs significantly less than its commercial counterpart. In this work, we fully characterize the proposed system and we demonstrate its performance for continuous gas monitoring and analyze different parameters such as response time, SC tunability, repeatability, selectivity, and sensitivity. The proposed system also can detect very wide-range of concentrations showing linear response to concentrations as high as 100% for CH$_4$ and 80% (8x 10$^5$ ppm) for NH$_3$.

II. SC LASER FOR AMMONIA AND METHANE DETECTION

The excitation laser used for the experiment is a relatively easy-to-build fiber-coupled, in-house made SC laser. The SC laser uses a directly modulated telecom-range diode laser-based amplifier (MLT-PLR-OMEM20, Manlight) operating at ~1550 nm as a pump laser with 3.2 ns pulse length duration. The pump laser is driven by an external pulse generator (TG2000, AIM-TTi), which is tunable from 30 kHz to 100 kHz. This tunability is vital for our application because the peak power is consequential in determining the broadening of the SC spectrum. The pulses from the laser are used to pump a standard dispersion-shifted fiber (DSF) (DCF4, Thorlabs) with a ZDW close to 1550 nm; this means our pump is very close to the ZDW, which is essential for standard SC generation [35]. The fiber output of the pump laser is directly spliced using a commercial splicer (FSM-100P+, Fujikura) on to the DSF so as to maximize the coupling efficiency and also for the ease of handling. In order to avoid back reflections from the end facet of the output fiber, the output fiber was angle cleaved - setting a cleaving angle of about 8 degrees in the tension cleaver (CT-100, Fujikura).

The angle-cleaved end of the fiber is placed in the ferrule inside the gas-cell shown in Figure 2. Details of the experimental setup are explained in the next section III.

The output spectrum of the SC is tailored to match the absorption bands of the species by switching between two settings. For NH$_3$ detection, we use a repetition rate of 30 kHz and a seed current of 1700 mA to increase the power spectral density (PSD) below the pump - since the region of interest (as seen in Figures 1(a) and 1(b)) is from about 1480 nm to 1550 nm. For CH$_4$ detection, we use a repetition rate of 91 kHz and a seed current of 3200 mA, to increase the PSD above 1610 nm - since this is the region of interest for CH$_4$ (as seen in Figures 1(a) and 1(c)).

III. EXPERIMENTAL SET-UP FOR ALL-FIBER SYSTEM FOR GAS SENSING

The applicability of SC laser described in the previous section is demonstrated for multi-species gas sensing. The angle-cleaved output fiber was placed inside a ceramic ferrule, such that the tip of the fiber is barely extruding out of the ferrules’ borehole. Then the other end of the ferrule is glued with an epoxy glue. Another ferrule of same dimensions is used for the HCPBG fiber, here, the facet of the fiber is flat-
cleaved because there is no issue of back reflection since the core is hollow (air). The HCPBG fiber is fixed into the ferrule similarly with epoxy; the two connectorized fibers are then mated with a mating-sleeve connector (Thorlabs, ADAF1). The mechanical splice is adjusted carefully to obtain high coupling efficiency between the two fibers while still maintaining some gap between the two ends to facilitate gas diffusion into the HCPBG fiber. This procedure is done carefully under optical microscope for higher precision. We found that a gap of ~80 µm between the SMF and the HCPBG fiber gave a coupling efficiency of ~65% while maintaining sufficient path for gas diffusion. The procedure is done again at the other end of the HCPBG fiber, where an optical patch cable is mechanically spliced to the HCPBG and then connected to either an Optical spectrum analyzer (OSA) (Assetrelay, ANDO AQ6317B) or a power meter (Thorlabs, S407C) for spectral characterization and power monitoring, respectively.

**IV. Multi-gas Detection**

**A. Spectral Analysis**

The output spectrum of the fiber is measured using an ANDO OSA with a resolution of 0.1 nm. Firstly, the entire chamber was purged with N₂ gas, and the background spectrum is recorded, shown in red in Figs. 3b and 4b. Then either NH₃ or CH₄ is passed through the chamber. The laser parameters were tuned to increase the power in the respective spectral regions as detailed in Section II and the spectrum was recorded. In the case of NH₃, the spectral measurement was done at several concentrations and in all cases the spectral absorption dips are clearly visible in the 1500-1550 nm band, as seen in Fig. 3b. We calculated the absorbance, log(I/I₀), of the gases; where I₀ and I is the transmission with pure N₂ and target gas respectively. This is depicted in Fig. 3a, where the measured absorbance perfectly captures the absorption band from HITRAN (with intensity cut-off of 1x10⁻²⁸ cm/mol). In particular, it accurately captures the strongest peak at 1531 nm which has been reported in many studies of ammonia near-IR spectroscopy [37], [38].
strongest peak at 1666 nm as shown in Fig. 4a. For both NH$_3$ and CH$_4$, however, each and every individual absorption line cannot be fully resolved because the actual absorption lines are much narrower than the maximum resolution of 0.1 nm of the OSA used. Therefore, in order to quantify the relative change in transmitted light between N$_2$ and any other gas mixture, one could calculate the area under the curve (spectrum) and compute the differences. In other words, we can alternatively monitor the changes in output power using a power meter. This will also permit for real-time continuous measurement [39].

\[ \text{Absorbance [a.u.]} \]

\[ \begin{align*}
\text{Wavelength [nm]} & \quad 1500 & \quad 1550 & \quad 1600 & \quad 1650 & \quad 1700 \\
\text{CH$_4$ absorption lines (Hitran)} & \quad \text{Experimental} & \\
\end{align*} \]

\[ \text{Absorbance [a.u.]} \]

\[ \begin{align*}
\text{Wavelength [nm]} & \quad 1500 & \quad 1550 & \quad 1600 & \quad 1650 & \quad 1700 \\
\text{CH$_4$ absorption lines (Hitran)} & \quad \text{Experimental} & \\
\end{align*} \]

B. Power Analysis

To have a continuous measurement of the gas in the system, a thermopile IR power meter (Thorlabs, S470C) was deployed, and the power is monitored in real-time. The power remained constant when pure N$_2$ is continuously flushed through the system. As seen in Fig. 5, after each NH$_3$ mixture, pure N$_2$ is passed into the system, and the output power always recovers to the initial position, showing the reliability of the system. When a 5% NH$_3$ mixture was injected, the power dropped and saturated at a specific value. When a higher concentration of NH$_3$ was used, the power drops to an even lower level. That is to say, we monitored the changes in the transmitted light (output power) at various concentrations of NH$_3$. The inset in Fig. 5 shows a linear fit to the power response, calculated as the percentage reduction in transmitted power, with the shaded region showing a 95% confidence band.

\[ \text{Total output power [mW]} \]

\[ \begin{align*}
\text{Time [minutes]} & \quad 10 & \quad 20 & \quad 30 & \quad 40 & \quad 50 & \quad 60 & \quad 70 & \quad 80 & \quad 90 & \quad 100 & \quad 110 & \quad 120 \\
0\% \text{ Ammonia} & \quad 21 & \quad 18 & \quad 15 & \quad 12 & \quad 9 & \quad 6 & \quad 3 & \quad 0 & \quad 0 & \quad 0 & \quad 0 & \quad 0 \\
5\% & \quad 20 & \quad 18 & \quad 16 & \quad 14 & \quad 12 & \quad 10 & \quad 8 & \quad 6 & \quad 4 & \quad 2 & \quad 0 & \quad 0 \\
10\% & \quad 21 & \quad 19 & \quad 17 & \quad 15 & \quad 13 & \quad 11 & \quad 9 & \quad 7 & \quad 5 & \quad 3 & \quad 1 & \quad 0 \\
20\% & \quad 22 & \quad 20 & \quad 18 & \quad 16 & \quad 14 & \quad 12 & \quad 10 & \quad 8 & \quad 6 & \quad 4 & \quad 2 & \quad 0 \\
33\% & \quad 23 & \quad 21 & \quad 19 & \quad 17 & \quad 15 & \quad 13 & \quad 11 & \quad 9 & \quad 7 & \quad 5 & \quad 3 & \quad 1 \\
67\% & \quad 24 & \quad 22 & \quad 20 & \quad 18 & \quad 16 & \quad 14 & \quad 12 & \quad 10 & \quad 8 & \quad 6 & \quad 4 & \quad 2 \\
80\% & \quad 25 & \quad 23 & \quad 21 & \quad 19 & \quad 17 & \quad 15 & \quad 13 & \quad 11 & \quad 9 & \quad 7 & \quad 5 & \quad 3 \\
\end{align*} \]

\[ \text{Ammonia Concentration [%]} \]

\[ \begin{align*}
\text{Signal response [%]} & \quad 0 & \quad 10 & \quad 20 & \quad 30 & \quad 40 & \quad 50 & \quad 60 \\
\text{Mean response} & \quad \text{Linear fit of response} & \quad \text{95\% Confidence band} & \\
\end{align*} \]

Fig. 5. (a) Continuous power log of NH$_3$ gas sensing: diluted concentrations from 5% to 80%. N$_2$ gas is passed into the system after each NH$_3$ mixture is flushed through the fiber. At 0% NH$_3$, only N$_2$ is in the fiber core. Red bars show the saturated power level for various concentrations and span in time, used in calculating signal response in (b). (b) Shows signal response of NH$_3$ at different concentrations, with the shaded area showing a 95% confidence band. Error bars in red.

A similar procedure was followed for CH$_4$. N$_2$ is then mixed with CH$_4$ from 0% to 100%. The continuous power log is shown in Fig. 6, and the relative decay in power (signal response) is plotted with the purple shaded area showing the 95% confidence band in Fig. 6(b). Similar to Fig. 5(b), the response is defined as the relative decay in power from the level for pure N$_2$ (in this case 9.2 mW), the saturated region for each concentration is shaded in red.
A gas mixture of \( \text{N}_2 \) and \( \text{CH}_4 \) (Air Liquide A/S) was used. Spectrum in black is \( \text{N}_2 \) and dotted red shows the 0.01\% \( \text{NH}_3 \) spectrum.

**B. Noise and Detection limit**

Although SC lasers are known for their pulse to pulse intensity fluctuations [41], the effect of the pulse to pulse fluctuations is significantly reduced when data is taken from the average of many pulses [2]. In this paper, the response from both the OSA and power meter sensor is averaged over \(~15,000\) pulses, which significantly increases the signal to noise ratio. With an OSA integration time of 1 second, the \( \text{NH}_3 \) and \( \text{CH}_4 \) spectra will be an average over a total of \( \text{30,000} \) and \( \text{91,000} \) pulses, respectively, due to the different repetition rates used in the two settings.

We compute the standard deviation (STD) of power log from the laser (after the mechanical splices) for 4 minutes- duration for half-cycle measurement (figure 8(a)). The STD for short-term stability is \( 1.57 \times 10^{-5} \). Which is similar to our long term stability for 6 hours. The Allan-werle deviation of the system for 6 hours stability measurement shows a theoretical detectable limit of 4 ppm as shown in Fig. 8(c).

Other sources of noise, such as white noise from the detector, room temperature fluctuations, etc. were not considered.

A gas mixture of \( \text{N}_2 \) and 0.01\% \( \text{NH}_3 \) was detected using the proposed system. The power meter reading did not show a noticeable difference when the entire spectrum is recorded, however, when a 1530 nm band-pass filter was
deployed at the output, a distinct difference in amplitude could be observed between the background and 0.01% NH3 (Fig. 7(b)) signal. Thus, the addition of filters could further improve the sensitivity, but not without compromising the all-fiberized system. Therefore the gas detection and monitoring was performed without any bulk components such as optical filters.

![Image](image_url)

Fig. 8. (a) Shows the stability of laser over 4 minutes. (b) Measurement by repeatedly passing of 50 % NH3 and recovery after passing N2. The demonstration of 2 and a half cycles shown in figure 8(b) was carried with a 50% NH3 mixture. (c) The Allan-Werle plot for ammonia sensing, showing the calculated detection limit, with optimal averaging time of 100s.

C. Repeatability

For any sensor, reliability is imperative; i.e., it must give consistent results when being used repeatedly. The proposed system is highly repeatable. We demonstrate this by taking a cycle of measurements for a fixed concentration and N2. The signal response of the gas always returns to a fixed value at given. Unlike metal oxides, there is no saturation of catalytic residue. Figure 8(b) shows a response measurement for 2 and a half cycles of the system while passing 50 % concentration of NH3 mixture followed by N2. It can be seen that the sensor shows consistent characteristics, thereby showcasing its repeatability.

VI. CONCLUSION

A compact and all-fiber gas sensor for multi-species gas detection based on readily available and off-the-shelf optical components is proposed. The all-fiber gas sensor is based on a simple home-built SC laser, which is cheap compared to commercially available SC sources. By tuning the peak power of the pump laser, the output spectrum of the SC is tailored to target the absorption features of two different gases within the telecommunication regime.

The results demonstrated here shows the possibility of targeting gasses such as NH3 and CH4, which are of great importance in the agricultural and farming sector. The proposed system is highly reliable for continuous monitoring, selective, cheap and sensitive to concentrations as low as 4 ppm when estimated using Allan-werle analysis. Effort can be given to further miniaturize the system by devising simple exhaust points on the gas-cell to pass ambient air for real-world applications; such will further improve the response time to less than a minute.

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