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Gas Sensor Based on Silicon Nitride Integrated Long Period Grating

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Abstract—In this paper, we demonstrate the potential of silicon nitride integrated long period grating (LPG) for gas sensing applications. The proposed LPG relies on the coupling of the single core mode propagating in the silicon nitride layer to cladding mode(s) by using a periodic sinusoidal width-modulation of the LPG core as pattern. This LPG is then used as CO₂ sensor. Here, as a proof of concept, the cladding layer is itself a porous medium functionalized to CO₂ using Polyhexamethylene Biguanide polymer. Preliminary experimental results show a constant sensitivity of 6.3 fm/ppm over CO₂ concentration ranging from [0-20%].

Index Terms—long period grating; gas sensing; silicon nitride; integrated photonics

I. INTRODUCTION

Providing a healthier and cleaner environment is one of the greatest challenge of the XXIst century that must be undertaken to improve people's well being as well as to protect Earth's environment and climate. As a result, it is of utmost importance to monitor, detect and quantify low concentration of certain gases either related to greenhouse effects such as carbon dioxide (CO₂) or methane (CH₄), or related to indoor air quality such as benzene, toluene or other volatile organic compounds.

For approximately a decade, integrated photonics has been intensely investigated for the development of highly sensitive lab-on-chip sensor applications. Integrated optical chemical sensors typically detect and measure the concentration of analytes via the interaction of the optical field with the surrounding environment, for example using the evanescent part of the mode field. For gas sensing applications, two main approaches are regarded as promising: absorption-based (spectroscopy) and refractometry-based mechanisms. While the former approach can achieve very high sensitivity without any functionalization layer, it relies on specific spectral signature for each gas molecule, which is usually located in the mid-infrared (MIR) spectral range (above 3 μm). Consequently, this spectroscopic approach requires both MIR light sources and low-loss materials at these wavelengths such as chalcogenide instead of silicon. On the other hand, refractometric sensors are based on the measurement of the refractive index unit

(RIU) change of a sensed region (e.g. top surface of a photonic chip via evanescent wave) induced by the presence or variation in concentration of a gas species [1], [2]. Contrary to spectroscopic sensors, these sensors require a chemo-optical transducer that has both gas-permeability and chemical affinity to the targeted gas molecule. Nevertheless, these sensors are not constrained by the use of a particular operating wavelength and can thus benefit from a wider range of fabrication technologies, photo-emitters and detectors [3]. As a result, various designs for integrated refractometric measurements have been proposed over the years, including integrated interferometers [4], ring resonators [5], plasmonics [6], photonic crystals [7] and gratings [8].

In this work, we propose to use the planar multimode Long Period Grating (LPG) described in [9] as gas sensing refractometer. The integrated LPG operates on wavelength-selective coupling of two or more co-propagative modes between the core and cladding modes whose effective indices (EI) can be analyzed using the resonance spectral features. This LPG structure consists in a sinusoidal longitudinal modulation of the waveguide's width to allow selective optical mode resonance with a mode contained in the cladding region. To enhance the LPG's sensitivity, the cladding layer is replaced by a porous medium functionalized to CO₂ using Polyhexamethylene Biguanide polymer (PHMB). Preliminary experimental results show a constant sensitivity of 6.3 fm/ppm over CO₂ concentration ranging from [0-20%].

This paper is organized in two main parts. First, in Section II, an overview of the LPG is presented. In Section III, the experimental setup and results are shown followed by a conclusion in section IV.

II. LPG DESIGN

A. LPG principles

LPGs have been successfully exploited in fibers due to their intrinsic capability for coupling energy to outer cladding modes, thereby allowing the optical power to substantially interact with the surrounding external environment. As shown in [10], [11], a phase-matching condition between two copropagative optical modes must be fulfilled to enable resonance to result in an exchange of energy. This condition can be derived

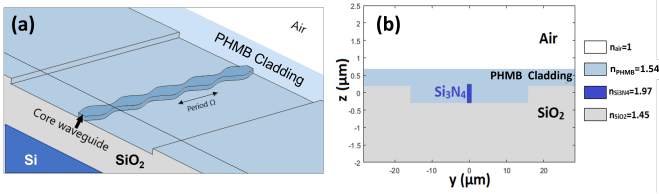


Fig. 1. (a) Illustration of the SiN LPG functionalized with PHMB with (b) its cross section material and RI profile.

from the Coupled Mode Theory (CMT) [12] and expressed as follows:

$$n_{eff,j} - n_{eff,l} = \frac{\lambda_{res}}{\Omega}, \quad (1)$$

where $n_{eff,j}$ and $n_{eff,l}$ are the EIs of the two coupled modes j and l , respectively, λ_{res} the center resonant wavelength and Ω the grating period.

The rate of energy exchange between the core and cladding modes upon resonance mainly depends on the modulation amplitude and optical field superposition at the modulated guide interfaces, as previously detailed in [9]. In refractometric sensing schemes employing LPGs, the presence of the analyte generally modifies the refractive index (RI) of the equivalent sensing layer which, in turn, modifies the propagation constants of the modes propagating in the waveguide. The difference in EI variation between the two coupled modes leads to a spectral shift of the optical resonance. This spectral shift can in turn be measured to determine the RI variations of the cladding layer itself. By considering (1), the sensitivity of the LPG can thus be expressed as:

$$S = \frac{\partial \lambda_{res}}{\partial n_{sens}} = \lambda \frac{\frac{\partial n_{eff,j}}{\partial n_{sens}} - \frac{\partial n_{eff,l}}{\partial n_{sens}}}{n_{g,j} - n_{g,l}}, \quad (2)$$

where $n_{g,x}$ is the group index of the x^{th} mode and n_{sens} the RI of the sensing layer of the mode x . Based on (2) and as shown in [9], the LPG sensitivity is enhanced by improving the modal overlap of the modes to couple in the sensing region, which can substantially increase $\frac{\partial n_{eff,x}}{\partial n_{sens}}$. This is achieved by inducing one of the modes to directly propagate in the sensing region, which is particularly applicable to gas sensors using porous cladding layers.

B. LPG structure and CO₂ functionalization

The LPG architecture consists of a Silicon Nitride (Si₃N₄) sinusoidally width-modulated integrated strip waveguide surrounded by a multimode cladding region, as shown in Fig. 1. As previously mentioned, the cladding layer is used as the sensing layer functionalized to CO₂, thus potentially increasing the coefficient $\frac{\partial n_{eff,cladding}}{\partial n_{sens}}$ to nearly 1. PHMB is considered here as the polymer host as it has been previously successfully demonstrated in photonic CO₂ sensing designs [13]–[15]. In addition, PHMB, soluble in water, is a low-toxicity polymer that is typically used as disinfectant and anti-septic. The high affinity of the polymer to CO₂ makes it a candidate for selector-free gas sensing. Additionally, the RI of PHMB at 1550 nm is ≈ 1.54 [13], which enables mode propagation on a silicon oxide substrate.

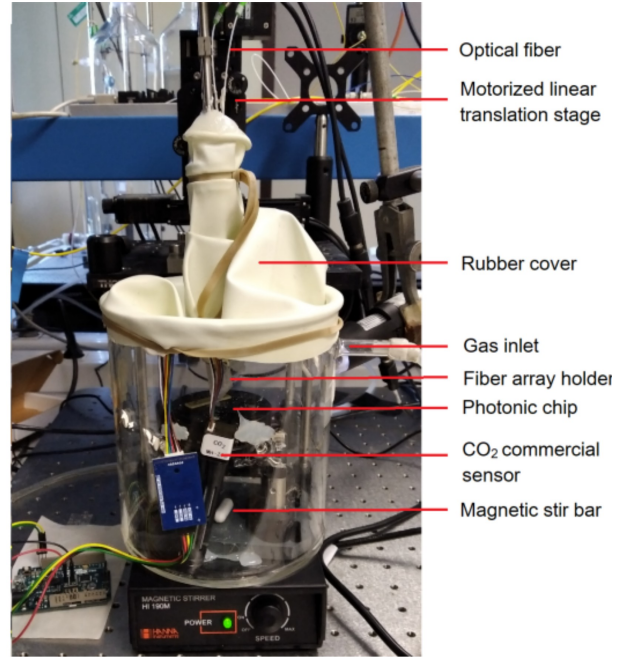


Fig. 2. Experimental setup of the CO₂ gas sensor.

The PHMB material is procured as a white powder with 95% purity from JHSS China. Methanol is hereby selected as solvent for its higher surface adhesion to silicon oxide and higher chemical volatility compared to water. The PHMB powder is dissolved with a ratio of 50 mg/mL of methanol and mixed using a magnetic stirrer for 3 hours. A 1.4 μm thin film is next deposited by spin coating onto a silanized $2 \times 2 \text{ cm}^2$ photonic chip at a spin rate of 800 rpm. The solvent is next evaporated by annealing at 90°C for 30 minutes.

III. EXPERIMENTAL RESULTS

A. Experimental Setup for Gas Sensing

To introduce the gas, a glass chamber is used as an impermeable container to allow the control of the gaseous environment into which the photonic chip is immersed, as shown in Fig. 2. The chamber has three openings: one gas inlet and outlet, respectively, and the third to allow the alignment of the fiber array with the grating couplers of the photonic chip to facilitate light injection and measurements. The third opening is sealed using a flexible rubber cover and silicone (as airtight seal) to prevent gas leakage and/or contamination of the gas chamber while enabling fiber array displacements. A commercial CO₂ sensor (MH-Z16) is also inserted into the chamber as reference. Finally, the chamber is placed on top of the magnetic stirrer regulator (stirrer placed inside the gas chamber) to ensure a better homogeneity of the gaseous environment within the chamber itself.

B. Preliminary CO₂ Sensing Results

To detect CO₂, the LPG sensor is positioned at least 2 mm away from the grating couplers on the layout (Fig. 3), thereby allowing the fiber array to be in full contact with the chip without obstructing the interaction of the gas with PHMB

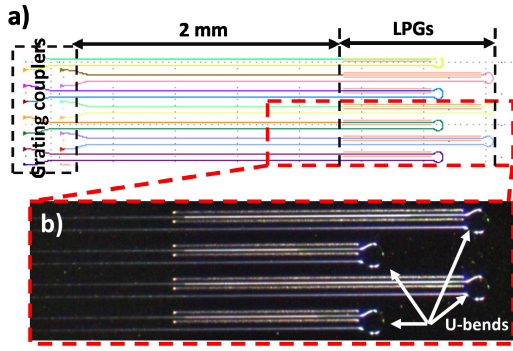


Fig. 3. a) layout of the SiN LPGs showing the grating couplers, LPG and U-bend. b) inset zoom of the fabricated SiN LPGs.

in the sensing region. The grating pairs, positioned at both ends of the U-bend, are joined by waveguides in which the fundamental mode propagates up to and through the LPG sensing region.

Consequently, for the analysis of the LPG's output signal, the superposed contributions from the grating couplers and waveguides are first de-embedded from the total output. A normalized coupling spectrum is obtained. Lorentzian curve fitting is applied to the de-embedded response.

Initial gas sensing tests have been performed by using the low-resolution fast scanning mode of the optical spectrum analyzer, with each scan carried out iteratively every 15 seconds. The measured spectra are analyzed in real-time with a dedicated LabVIEW program. Two experimental barriers are then observed during the measurements:

- 1) During gas-flow opening, the applied gas pressure is found to lead to an instantaneous virtual spectral shift of the resonance. This could be attributed to a minor variation of grating coupling behavior induced by minor vibrations of the grating coupler upon initial gas flow.
- 2) The reference CO₂ sensor exhibits an inertia during the measurement, in part due to its small dimension compared to the relatively large volume of the gas enclosure. Depending on the concentration injected, the gas mixture or atmosphere is estimated to generally stabilize or homogenize after a few minutes. Upon introduction of CO₂ into the chamber, the resonance shifts before stabilization of the gaseous atmosphere (i.e. after a few minutes) can be observed in Fig. 4 (a) highlighted as gas flow area. The CO₂ concentration measured by the reference sensor after stabilization of the atmosphere in the chamber is displayed a few minutes after gas flow.

To compensate the perturbations induced during measurements by gas surge immediately after valve opening and the relatively slow stabilization of the gaseous atmosphere in the chamber, the measurements have been performed over 1h30 at very low gas flow. Here, high-resolution low-speed scans are instead performed and analyzed against reference measurements. A quasilinear spectral shift is observed, as illustrated in Fig. 4(b). Linear interpolation is then applied on the measured data to obtain a sensitivity of 6.8 fm/ppm. Subsequently, based on the estimated LPG bulk sensitivity

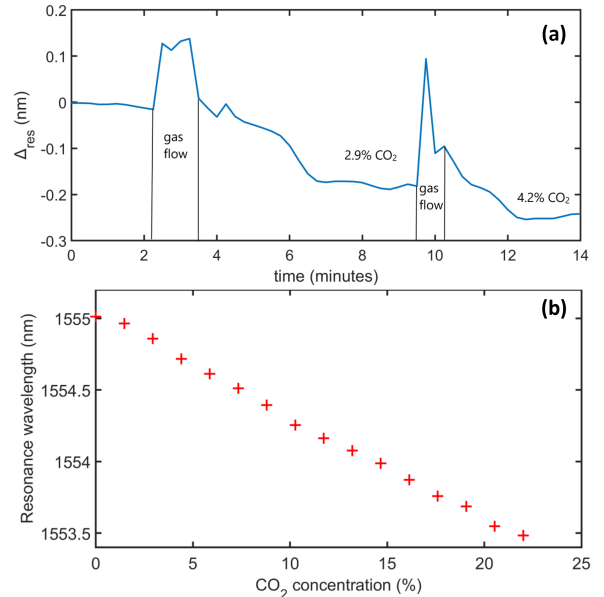


Fig. 4. Preliminary results from CO₂ sensing experiment in enclosure under (a) low-resolution fast-spectral scanning mode and (b) high resolution slow-spectral scanning mode.

[9], the RI sensitivity of the PHMB layer is estimated to be $\approx 4.7 \cdot 10^{-9}$ RIU/ppm, corroborating the accepted value of $\approx 6 \cdot 10^{-9}$ RIU/ppm reported in the literature [13]. The discrepancy between these two sensitivity values can be attributed to different coating methods or presence of chemical impurities, thus giving rise to slightly different solubility characteristics.

IV. CONCLUSION

We have presented an integrated LPG sensor architecture functionalized to CO₂ with a PHMB layer. This architecture achieves a sensitivity of 6.8 fm/ppm of CO₂ which corroborates with the PHMB RI sensitivity. As a result, in the future, LPG with higher sensitivity such as the one described in [16] can be used to achieve even higher sensitivity close to 32 fm/ppm. This type of design thus appears promising and can benefit from improvements or hybridations, potentially involving new sensing mechanisms, like plasmonics or photonic crystals.

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REFERENCES

- [1] M. Ingvaldsen, "Sensitivity to pressure and methane of a cryptophane-a doped polymer," Master's thesis, UiT Norges arktiske universitet, 2015.
- [2] A. Urrutia, I. Del Villar, P. Zubiate, and C. R. Zamarréño, "A comprehensive review of optical fiber refractometers: Toward a standard comparative criterion," *Laser & Photonics Reviews*, vol. 13, no. 11, p. 1900094, 2019.
- [3] E. Luan, H. Shoman, D. M. Ratner, K. C. Cheung, and L. Chrostowski, "Silicon photonic biosensors using label-free detection," *Sensors*, vol. 18, no. 10, p. 3519, 2018.
- [4] F. T. Dullo, S. Lindecrantz, J. Jágorská, J. H. Hansen, M. Engqvist, S. A. Solbø, and O. G. Hellesø, "Sensitive on-chip methane detection with a cryptophane-a cladded mach-zehnder interferometer," *Opt. Express*, vol. 23, pp. 31564–31573, Nov 2015.
- [5] N. A. Yebo, S. P. Sree, E. Levrau, C. Detavernier, Z. Hens, J. A. Martens, and R. Baets, "Selective and reversible ammonia gas detection with nanoporous film functionalized silicon photonic micro-ring resonator," *Opt. Express*, vol. 20, pp. 11855–11862, May 2012.
- [6] A. Zaki, K. Kirah, and M. Swillam, "Integrated optical sensor using hybrid plasmonics for lab on chip applications," *Journal of Optics*, vol. 18, p. 085803, 08 2016.
- [7] M. Danaie and B. Kiani, "Design of a label-free photonic crystal refractive index sensor for biomedical applications," *Photonics and Nanostructures - Fundamentals and Applications*, vol. 31, pp. 89 – 98, 2018.
- [8] S. Jain, S. Srivastava, S. Rajput, L. Singh, P. Tiwari, A. K. Srivastava, and M. Kumar, "Thermally stable optical filtering using silicon-based comb-like asymmetric grating for sensing applications," *IEEE Sensors Journal*, vol. 20, no. 7, pp. 3529–3535, 2020.
- [9] C. Deleau, H. C. Seat, H. Tap, F. Surre, and O. D. Bernal, "Integrated width-modulated sin long period grating designed for refractometric applications," *Journal of Lightwave Technology*, pp. 1–1, 2021.
- [10] L. Jin, W. Jin, J. Ju, and Y. Wang, "Coupled local-mode theory for strongly modulated long period gratings," *Journal of Lightwave Technology*, vol. 28, no. 12, pp. 1745–1751, 2010.
- [11] H. F. Taylor and A. Yariv, "Guided wave optics," *Proceedings of the IEEE*, vol. 62, no. 8, pp. 1044–1060, 1974.
- [12] A. W. Snyder and J. Love, *Optical waveguide theory*. Springer Science & Business Media, 2012.
- [13] G. Mi, C. Horvath, M. Aktary, and V. Van, "Silicon microring refractometric sensor for atmospheric CO₂ gas monitoring," *Optics express*, vol. 24, no. 2, pp. 1773–1780, 2016.
- [14] N. L. Kazanskiy, M. A. Butt, and S. N. Khonina, "Carbon dioxide gas sensor based on polyhexamethylene biguanide polymer deposited on silicon nano-cylinders metasurface," *Sensors*, vol. 21, no. 2, p. 378, 2021.
- [15] W. Ma, J. Xing, R. Wang, Q. Rong, W. Zhang, Y. Li, J. Zhang, and X. Qiao, "Optical fiber fabry-perot interferometric CO₂ gas sensor using guanidine derivative polymer functionalized layer," *IEEE Sensors Journal*, vol. 18, no. 5, pp. 1924–1929, 2018.
- [16] C. Deleau, H. C. Seat, O. Bernal, F. Surre, *et al.*, "High-sensitivity integrated sin rib-waveguide long period grating refractometer," *Photonics Research*, vol. 10, no. 2, p. 02000564, 2022.