Carbon dioxide absorption spectroscopy with a mid-infrared silicon photonic waveguide

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Carbon dioxide is a vital gas for life on Earth, a waste product of human activities, and widely used in agriculture and industry. Its accurate sensing is therefore of great interest. Optical sensors exploiting the mid-infrared light absorption of CO$_2$ provide high selectivity, but their large size and high cost limit their use. Here, we demonstrate CO$_2$ gas sensing at 4.2 µm wavelength using an integrated silicon waveguide, featuring a sensitivity to CO$_2$ of 44% that of free-space sensing. The suspended waveguide is fabricated on a silicon-on-insulator substrate by a single-lithography-step process, and we route it into a mid-infrared photonic circuit for on-chip-referenced gas measurements. Its demonstrated performance and its simple and scalable fabrication make our waveguide ideal for integration in miniaturized CO$_2$ sensors for distributed environmental monitoring, personal safety, medical, and high-volume consumer applications.
accelerating the development of on-chip optical CO$_2$ sensors.

To be the core element of miniaturized optical CO$_2$ sensors, photonic waveguides must fulfill two key requirements. They must support light modes at 4.2 $\mu$m wavelength with a large portion of the field outside the waveguide core material, to enable interaction with the gas, and they should have a low base propagation loss, as this limits the applicable waveguide length. The ratio between these characteristics fully determines the sensing performance of the waveguide, and is expressed by the figure of merit $FOM = \Gamma/\alpha$, introduced by Kita et al. [19]. Here, $\alpha$ is the waveguide base attenuation coefficient, including all losses not due to CO$_2$ absorption, such as material, scattering, curvature, and substrate losses, $\Gamma = \partial n_{\text{mode}}/\partial n_{\text{clad}}$, where $n_{\text{mode}}$ is the light mode’s effective index, is the external confinement factor expressing the waveguide’s sensitivity to changes in the cladding’s refractive index $n_{\text{clad}}$ [19–21]. Contrarily to the evanescent field ratio (EFR), i.e. the portion of optical power propagating outside the waveguide core, $\Gamma$ correctly describes the sensitivity of any waveguide, including those with high core-cladding refractive index contrast [21]. The FOM, in conjunction with the waveguide length and the signal-to-noise ratio of the measurement setup, determines the achievable limit of detection of the system.

A variety of integrated waveguides for the mid-IR have been presented [12, 13, 16, 18]. A particularly attractive waveguide material is silicon (Si), because it combines a large transparency wavelength (Fig. 2 (d)) has an EFR of 65.4% and an external confinement factor $\Gamma$ of 52.0%. The waveguide circuit was fabricated on a commercial silicon-on-insulator (SOI) substrate with a 220 nm Si device layer and a 3 $\mu$m SiO$_2$ buried oxide (BOX) layer by a single electron-beam lithography step, dry etching of the Si, wet etching of the SiO$_2$, and cleaving. We used electron-beam lithography because of its rapid turnaround time, but the minimum feature size in the lithography step, dry etching of the Si, wet etching of the SiO$_2$, and cleaving. We used electron-beam lithography because of its rapid turnaround time, but the minimum feature size in the circuit is compatible with stepper photolithography.

We characterized the CO$_2$ sensing performance of our photonic waveguide with the setup shown in Fig. 2 (e). We focused 4.24 $\mu$m continuous-wave linearly polarized light from a distributed-feedback quantum cascade tunable laser with single-wavelength emission onto the input facet of the waveguide. We placed the waveguide chip inside a steel case with a mid-IR-transparent window that allowed the visualization of the chip surface by a mid-IR camera equipped with a cooled InSb-

![Fig. 2.](image)

(a), (b), (c) False-color SEM images of the fabricated Si photonic waveguide. (a) Waveguides branching out from a splitter. (b) A support structure where the Si waveguide was partially removed to expose the SiO$_2$ pillar. (c) A close-up view of the suspended waveguide. (d) Cross-sectional FEM simulation of the waveguide, displaying the electric field profile of the de-confined fundamental TE mode. (e) Schematic of the setup used for CO$_2$ sensing, including a sample mid-IR image of the operating waveguide.

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We placed the waveguide chip inside a steel case with a mid-IR-transparent window that allowed the visualization of the chip surface by a mid-IR camera equipped with a cooled InSb-
detector and a 1× macro lens. The chip case also had an inlet and an outlet that enabled controlled gas injection and a steady flow inside the case. The mid-IR camera aided the alignment of the waveguide input to the focused light for in-coupling and detected the output signal from the grating couplers. We alternately injected nitrogen (N\textsubscript{2}) and dilutions of CO\textsubscript{2} in N\textsubscript{2}, purchased pre-mixed, in concentrations of 0.1%, 0.5%, 1%, and 2.5% in 1 min intervals at a flow rate of 600 mL/min. For each CO\textsubscript{2} concentration, we repeated the three-minute N\textsubscript{2}-CO\textsubscript{2}-N\textsubscript{2} measurement at different wavelengths across the CO\textsubscript{2} absorption peak highlighted in the inset of Fig. 1. To reduce light absorption by the atmospheric CO\textsubscript{2} along the free-space path between the laser head and the focusing lens and between the chip case and the camera, we enclosed these sections in brass tubes and continuously flushed those with N\textsubscript{2}.

Fig. 3 shows the results of four sample measurements, one for each CO\textsubscript{2} concentration tested. The top panels show the intensity time traces of the three waveguide outputs, including all recorded frames. Here, the intensity drop during CO\textsubscript{2} injection results from light absorption along the entire light path, i.e. along the waveguide, including the input section before the first output, and the free-space sections in the setup, where residual ambient CO\textsubscript{2} is present despite the N\textsubscript{2} flushing. By extracting the decay rate of the light intensity along the waveguide from the three outputs, as shown in the insets of Fig. 3 for two sample frames, we continuously measured the waveguide propagation loss in a real-time cut-back measurement. We accounted for the splitting loss using a reference waveguide circuit with no additional waveguide length between splitters (Fig. 2 (e)). The measured propagation loss is displayed in the bottom panels of Fig. 3. The propagation loss during N\textsubscript{2} injection, i.e. the waveguide base loss, is 4.6 dB/cm. The increase in the loss during CO\textsubscript{2} injection, clearly visible at all tested concentrations, is the excess loss caused exclusively by absorption along the straight waveguide sections between grating outputs. Thus, the difference between the time-averaged propagation loss during N\textsubscript{2} injection and the one during CO\textsubscript{2} injection is the waveguide CO\textsubscript{2} absorption loss.

Fig. 4 shows the waveguide CO\textsubscript{2} absorption loss for all concentrations and wavelengths measured. We compare the measured waveguide CO\textsubscript{2} absorption loss with the predicted free-space CO\textsubscript{2} absorption loss at corresponding pressure and temperature, as listed in the HITRAN database [9] and confirmed by our free-space reference measurement. We find that the \( \Gamma \) of our waveguide, i.e. its sensitivity to CO\textsubscript{2}, is 44% that of free-space, a value close to the simulated 46.3%. The measured FOM of our waveguide is thus 0.42 cm. The standard deviation of the measured waveguide propagation loss in N\textsubscript{2} (Fig. 3) indicates that the smallest loss change measurable in our setup is 0.17 dB, corresponding to a CO\textsubscript{2} concentration of 330 ppm. According to the measured FOM, the optimal length [19, 23] for our waveguide to sense present-day atmospheric levels of CO\textsubscript{2}, i.e. 400 ppm, is 9 mm.

In Table 1, we compare our waveguide to other relevant integrated gas-sensing waveguides. Our waveguide features the highest theoretical and experimentally demonstrated \( \Gamma \) amongst all the listed waveguides, and exhibits the highest FOM amongst...
the mid-IR waveguides.

We note that Ranacher et al. [24, 25] measured a $\Gamma$ consistently higher than the simulated one. This might be caused by the absorption and subsequent release of CO$_2$ by the plastic tubing and chip case used in the experiments. Such memory effects are particularly relevant when injecting the CO$_2$ mixtures in order of decreasing concentration, and result in higher CO$_2$ levels than intended. Ultimately, the performance of these waveguides is limited by the high intrinsic mode loss caused by the mid-IR absorption of the SiO$_2$ cladding and large support structures.

Tombez et al. [22] achieved a high FOM in methane (CH$_4$) sensing by probing an overtone absorption band of CH$_4$ using the fundamental TM-polarized mode at 1.65 µm wavelength. For their waveguide design, and theoretically for all the listed waveguides, the TM mode is less confined than the TE mode, and requires increasing the spacing between the waveguide and the bottom cladding and large support structures.

We achieve a high $\Gamma$ with the fundamental TE mode thanks to the small waveguide thickness, and a potentially low base loss thanks to the almost complete removal of the 3 µm-thick SiO$_2$ BOX layer. The difference between the simulated and the measured base loss is due to sidewall roughness. The loss could be reduced with a higher-quality fabrication process, possibly including thermal oxidation and selective oxide etching to smoothen the waveguide surfaces. Since the etched sidewalls constitute less than one tenth of the waveguide surface, our design may achieve very low scattering losses. Moreover, optimization of the support structure design and an increase in their pitch could further reduce the theoretical intrinsic loss.

In conclusion, we have demonstrated the optical absorption spectroscopy of CO$_2$ concentrations down to 0.1 % using a 3.2 mm-long low-confinement Si photonic waveguide for 4.24 µm wavelength. The waveguide was fabricated on an SOI platform with a single lithography step. The waveguide, intrinsically broadband, can operate in conjunction with both broad- and narrow-band sources and detectors. It has a small footprint and can easily be routed to form mid-IR photonic circuits, potentially including components such as resonant cavities and spectral filters. By integrating MMI splitters, we implemented for the first time a waveguide circuit for on-chip-referenced gas measurements, to eliminate errors due to the ambient CO$_2$ and ensure that the characterized sensing performance is ascribable only to the waveguide. In this way, we have demonstrated that the $\Gamma$, and hence the sensitivity to gas, of our waveguide is 44 % that of free-space sensing, and its FOM is 0.42 cm. Compared to previous mid-IR gas-sensing waveguides, our waveguide exhibits the highest external confinement factor and a three-fold improved FOM. This demonstrated performance and the simple, cost-effective, and scalable fabrication make our integrated photonic waveguide ideal for mass production and large-scale adoption. It has the potential to become the choice component for an increasingly broad range of applications, such as portable and distributed environmental monitoring, and high-volume medical and consumer applications.

**FUNDING INFORMATION**

This work was partially funded by grants from VINNOVA (2016-02328 and 2017-05108), SLL (20150910), and SSF (GMIT14-0071).

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**Table 1. Comparison of reported gas-sensing waveguides.**

| Pol. | $\lambda$ [µm] | Gas     | Intrinsic mode loss $\Gamma [\text{cm}]$ | FOM $\%$ | Waveguide base loss $\alpha_{IB}$ [dB/cm] | FOM $\%$ |
|------|----------------|---------|----------------------------------------|---------|----------------------------------------|---------|
| TE   | 4.24           | CO$_2$  | 2.93                                   | 4.6     | 44                                     | 0.42    |
| TE   | 4.23           | CO$_2$  | 4.44                                   | 4.0†    | 14                                     | 0.15    |
| TM   | 1.65           | CH$_4$  | 1.73                                   | 2       | 25.4                                   | 0.55    |

*Assuming zero gas absorption, excluding scattering losses, and including substrate leakage losses and, for the suspended designs, losses due to the support structures.
† Measured at 4.17 µm wavelength.

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