Characterization of an MIR-LED as an excitation source for photoacoustic sensing of N_2O

Mila V. da Rocha[a,b], Daniel da S. Santos[a,c], Guilherme R. Lima[d], Leonardo Mota[c], Marcelo G. da Silva[c], and Marcus Wolff[a]

[a]Hamburg Universisty of Applied Sciences, Berliner Tor 21, 20099 Hamburg, Germany
[b]Instituto Federal Fluminense, Rua Cel. Walter Kramer, 357 - Parque Santo Antonio, Campos dos Goytacazes - RJ, 28080-565, Brazil
[c]Universidade Estadual do Norte Fluminense Darcy Ribeiro , Av. Alberto Lamego 2000, Campos dos Goytacazes, RJ, 28013-602, Brazil
[d]Universidade Federal do Espírito Santo, s/n○, Alegre, Espírito Santo, 29500-000,Brazil

ABSTRACT

Nitrous oxide (N_2O) is an extremely effective greenhouse gas emitted in large quantities from agriculturally used soils. Traditional methodologies to monitor this gas are typically associated with significant costs. In this study, we report on the characterization of a low-cost MIR-LED with spectral emission centered at 4.5 μm, to be used as excitation source in photoacoustic detection of nitrous oxide (N_2O). N_2O is emitted, in agriculture, in trace levels. Preliminary results suggest that the low power and divergence of the LED poses a significant challenge in the development of a LED based PA sensor, and that further developments on the PA cell are required to allow a better assessment of the feasibility of this light source to monitor N_2O in agricultural sites.

Keywords: MIR-LED, photoacoustic spectroscopy, nitrous oxide, greenhouse gas.

1. INTRODUCTION

Nitrous oxide (N_2O) is a greenhouse gas and it's concentration in the atmosphere has significantly increased over the past four decades, to which agricultural emissions contributed considerably.[1] Photoacoustic (PA) sensors have been used to monitor these emissions at trace levels in agricultural settings.[2,3] In PA detection, the beam quality of the excitation source is crucial to ensure low background signals and increased sensitivity. The ideal beam profile is collimated, with most of its power concentrated in a narrow waist, to enable good coupling with PA resonators. Therefore, lasers have traditionally been preferred as radiation source.

Quantum cascade laser and interband cascade lasers are options in the mid-infrared that are often used in photoacoustic sensors, due to their power in the tens of mW range and narrow spectral linewidth.[2,4] However, these devices are typically expensive, which ultimately hinders the development of cost-effective, portable sensors for real-time measurements in agricultural settings. Recent research efforts have focused on the use of light emitting diodes (LED) due to their significantly reduced cost.[3,5] Challenges of using an LED as excitation source include the highly diverging beam, its spectrally broad emission bandwidth and its comparatively low power in the mid-infrared spectral range. In this study, we report on the characterization and optimization of an MIR-LED to be used as excitation source in a photoacoustic spectroscopy setup to detect N_2O.

Further author information: (Send correspondence to Mila V. da Rocha.)
Mila V. da Rocha.: E-mail: mila.rocha@iff.edu.br, Telephone: + 49 17936227880

2. MIR-LED CHARACTERIZATION

The light-emitting diode (MIR-LED - Nanoplus GmbH), featuring an infrared spectral bandwidth (FWHM) of approximately 1 μm centered at 4.5 μm, matches a strong rotational-vibrational transition of N_2O, Fig. 1 (left). The LED was mounted in a heatsink (Nanoplus GmbH), the operating current was provided by a power supply (LDC402B, Profile) and the operating temperature was set by a temperature controller (TTC001, THORLABS).

Due to the divergent nature of the LED beam, the diode was coupled to an AR-coated aspheric lens (390037 - E, ThorLabs) of 5.5 mm diameter and 1.873 mm focal length. As the diode optical power was very low, effectively less than 1 mW, the lens was placed at a distance of 0.8 mm from the LED to maximize the collected light.

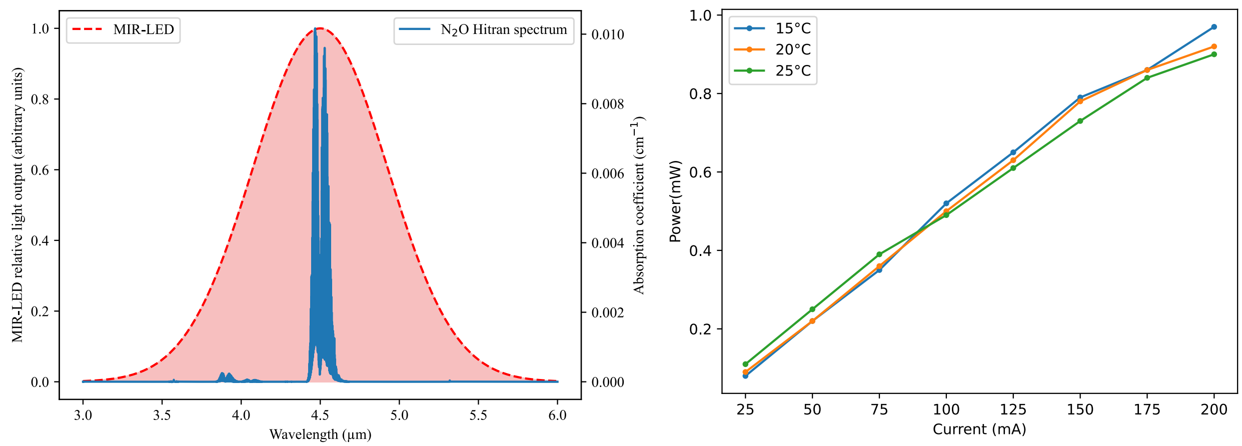

Figure 1. LEFT: MIR-LED spectral emission (red) and N_2O (100 ppm) absorption (blue) from the Hitran[6] database. RIGHT: Optical power response to varying parameters of current and temperature.

An initial step consisted of investigating the optical output response to varying parameters of operating current and operation temperature. In this measurement, a power meter (S401C, Thorlabs) was placed as close as possible to the LED and the results are displayed in Fig. 1 (right). Optimal performance was observed for the maximum current of 200 mA and operation temperature of 15°C, therefore these were the parameters applied in subsequent experiments.

A detailed investigation of the beam profile was carried out and light intensity was monitored by a PbSe preamplified photodetector (PDA20H-EC, Thorlabs), with 4mm^2 of active area, at different distances from the LED, Fig. 2. To ensure that the detected signal was free from interferences from the laboratory environment, the diode was modulated by a function generator (OWON AG1022) with a sinusoidal wave at the frequency of 3.785 kHz and peak to peak amplitude off 200mA. The signal from the photodetector was then amplified using a lock-in amplifier (7265 Signal Recovery) locked at the same reference frequency. The experimental setup also included a diode driver (ThorLabs TLD001) and a temperature controller (TTC001). The optical power distribution, in μW/mm^2[*], across a horizontal and a vertical plane can be observed in Fig. 3.

Considering the FWHM approach, the beam diameter at the shortest measured distance (1 mm) from the LED (z axis) can be estimated as approximately 5.9 mm in the x direction and 5.8 mm in the y direction, and the divergency was estimated as approximately 14 and 16 degrees in xz and yz planes, respectively. In addition, the intensity of the beam rapidly decreases along the x axis, yielding an effective optical beam length of approximately 35 mm[†].

[*] Unit obtained from a calibration of the photodetector with a power meter (3A-F-SH, OPHIR).

[†] Effective optical beam length: here defined as the distance along the axial direction over which the beam retains sufficient power and a Gaussian distribution, to remain optically useful.

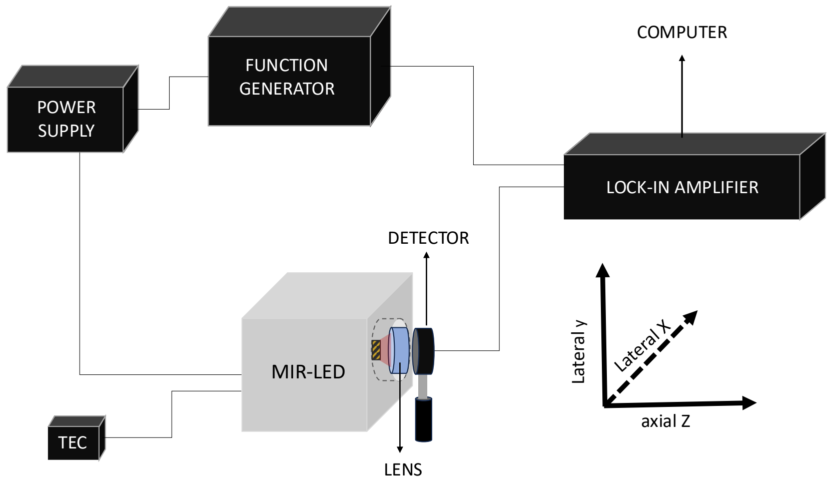

Figure 2. Experimental setup employed in the beam profile characterization.

3. PHOTOACOUSTIC SENSORING OUTLOOK

In a PA sensor, modulated radiation is absorbed by the molecule of interest, promoting it to an excited state. The energy is then released through non-radiative decay in the form of heat, producing periodic temperature fluctuations. A pressure wave is thus generated, with an amplitude proportional to the gas concentration.[7] In recent years, many photoacoustic cell designs have been proposed to improve the minimum detection limits of trace-level environmental gases.[8–11]

The analysis of the LED beam profile characteristics suggests that its divergent pattern, low power and short effective beam length would demand a PA cell that could compensate these limitations to provide detection limits in the trace level range. Recently, a T-shaped cell has shown good performance when combined with an LED, to detect NO_2 (nitrogen dioxide)in sub-ppm levels.[3]

4. CONCLUSION

LEDs have been used in the development of low-cost PA sensors to detect gases at trace levels and are, therefore, a promising choice in the development of similar sensors to monitor N_2O emissions in agricultural settings. In this study, a MIR-LED was characterized and an investigation of its beam profile suggests that an experimental photoacoustic setup could benefit from the implementation of a PA cell with a customized geometry to optimize the optical path and increase sensitivity. Future steps will, therefore, focus on further developing the system before assessing it's feasibility with natural samples.

ACKNOWLEDGMENTS

The authors acknowledge and appreciate the support of DAAD-CAPES (PROBRAL) and HAW Hamburg for fostering international cooperation between Brazil and Germany.

REFERENCES

[1] Tian, H., Pan, N., Thompson, R. L., Canadell, J. G., Suntharalingam, P., Regnier, P., and et al., "Global nitrous oxide budget (1980–2020)," *Earth System Science Data* **16**(6), 2543–2604 (2024).

[2] Lopes, L. A. S., Lopes, D. C. L., Mota, L., Lima, G. R., Alves, B. J. R., and da Silva, M. G., "Sensitive gas spectroscopy method for real-time determination of urease activity via ammonia production," *Analytical Methods* (2025). In press.

[3] Kapp, J., Weber, C., Schmitt, K., Pernau, H. F., and Wöllenstein, J., "Resonant photoacoustic spectroscopy of no_2 with a uv-led based sensor," *Sensors* **19**(3), 724 (2019).

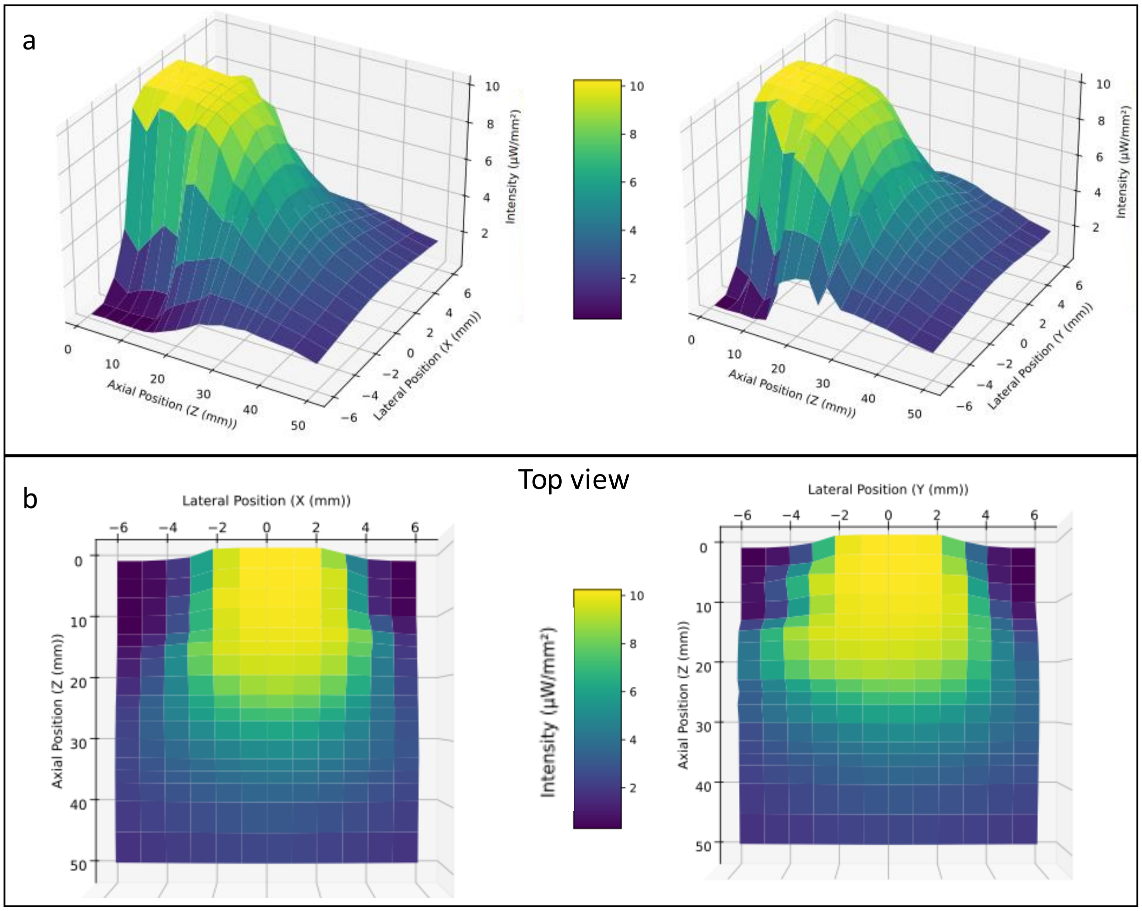

Figure 3. a) LED light intensity distribution across an xz plane (left) and an yz plane (right). b) Top view.

[4] Bahr, M.-S. and Wolff, M., "PAS-based analysis of natural gas samples," *Frontiers in Chemistry* **11**, 1328882 (2023).

[5] Rey, J. M. and Sigrist, M. W., "Non-dispersive sensing scheme based on mid-infrared led and differential mode excitation photoacoustic spectroscopy," *Photoacoustics* **29**, 100455 (2023).

[6] Gordon, I. E., Rothman, L. S., Hargreaves, e. R., Hashemi, R., Karlovets, E. V., Skinner, F., Conway, E. K., Hill, C., Kochanov, R. V., Tan, Y., et al., "The hitran2020 molecular spectroscopic database," *Journal of quantitative spectroscopy and radiative transfer* **277**, 107949 (2022).

[7] Miklós, A., Hess, P., and Bozóki, Z., "Application of acoustic resonators in photoacoustic trace gas analysis and metrology," *Review of Scientific Instruments* **72**(4), 1937–1955 (2001).

[8] Zhang, L., Liu, L., Zhang, X., Yin, X., Huan, H., Liu, H., Zhao, X., Ma, Y., and Shao, X., "T-type cell mediated photoacoustic spectroscopy for simultaneous detection of multi-component gases based on triple resonance modality," *Photoacoustics* **31**, 100492 (2023).

[9] Huang, Q., Wei, Y., and Li, J., "Simultaneous detection of multiple gases using multi-resonance photoacoustic spectroscopy," *Sensors and Actuators B: Chemical* **369**, 132234 (2022).

[10] Lima, G. R., Pelais, A., Neto, A. G., Coutinho, M. F., Esquef, I. A., Batista, A. G. T., and Mota, L., "Acoustic modeling and performance evaluation of 3d-printed and metal differential photoacoustic sensors for trace gas detection," *Measurement* **229**, 114422 (2024).

[11] Gong, Z., Gao, T., Mei, L., Chen, K., Chen, Y., Zhang, B., Peng, W., and Yu, Q., "Ppb-level detection of methane based on an optimized t-type photoacoustic cell and a nir diode laser," *Photoacoustics* **21**, 100216 (2021).