Mode-hop compensation for intracavity sensing via chip voltage in an external-cavity QCL

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Abstract

In this paper we describe a technique to perform intracavity gas sensing by detecting changes in the QCL voltage. The influence of mode-hops is compensated by a data acquisition and processing based on a dual wavelength scanning. This allows to perform gas detection over the full cavity spectral range (1277 cm^{-1}-1348 cm^{-1}) without the use of a mode-hop free setup. First results of measurement of the CH_4 absorption spectrum are presented.

1 Introduction

Nowadays, the development of gas detection systems is a well-established research field but still constantly evolving with technological advancements. It is found in a lot of applications for air quality monitoring, defense and security, medical analysis and more. To perform gas detection, a spectral region of interest is the Mid-Infrared (Mid-IR) due to the presence of the fundamental absorption bands of molecules. Compared to the Near-Infrared where there are harmonic bands, Mid-IR spectra are less complex and absorption bands are more intense. Thus, the potential of Mid-IR detection is easier and faster data processing as well as more efficient systems. In order to reach the Mid-IR region a lot of sources exist such as Optical Parametric Oscillators (Hemming et al., 2013; Budni et al., 2000; Lippert et al., 2010), fiber lasers (Seddon et al., 2010; Henderson-Sapir, Munch, and Ottaway, 2014; Woodward et al., 2019) and semiconductor lasers (Olafsen et al., 1998; Vurgaftman et al., 2009; Yao, Hoffman, and Gmachl, 2012; Faist, Capasso, Sirtori, et al., 1996; Lee et al., 2007; Felix et al., 1997; Ikyo et al., 2016) for example. The laser team in GSMA (Groupe de Spectrométrie Moléculaire et Atmosphérique) in Reims, France, is specialized in the development of gas detection devices and techniques based on Quantum Cascade Laser (QCL) (Grossel, Zéninari, Parvitte, Joly, Courtois, and Durry, 2008; Grouiez, Parvitte, et al., 2009; Grouiez, Zéninari, et al., 2010; Joly et al., 2011; Vallon et al., 2016; Bizet et al., 2017). Since the first realization of a QCL in 1994 (Faist, Capasso, Sivco, et al., 1994), this semiconductor laser has been used for diverse applications such as environmental gas detection (Maamary et al., 2016; Nelson et al., 2002), optical countermeasure (Maulini et al., 2009), explosive detection (Mukherjee, Von Der Porten, and Patel, 2010; Papantonakis et al., 2009), or in biomedical with breath analysis (Shorter et al., 2010; Marchenko et al., 2013; Reyes-Reyes et al., 2015), blood or serum analysis (Brandstetter et al., 2013; Blake Martin, Mirov, and Venugopalan, 2005) and imaging (Kröger et al., 2014; Kröger-Lui et al., 2015; Haase et al., 2015).

As for the gas detection technique, a great variety exist and we cannot list them all here. For example, we only mention a few of interest such as the multi-pass absorption technique (McManus, Kebabian, and Zahniser, 1995; Liu, Wang, et al., 2015), Cavity Ring Down Spectroscopy (Romanini et al., 1997; Berden, Peeters, and Meijer, 2000) and Intracavity Spectroscopy (Baev, Latz, and Toschek, 1999; Kachanov, Charvat, and Stoeckel, 1994). Each of them has its advantages and disadvantages leading to different performances and easy-to-setup systems. However, in most cases, the use of a photo-detector is necessary because the signal of interest is the light intensity. This leads to potential experimental problems due to detector alignment or light collection for example. Moreover, the spectral range of the detector must match the application. In most cases, both laser source and detector must be replaced to explore a new spectral range. Photothermal and photoacoustic spectroscopy are two solutions to the optical detector need. Photothermal spectroscopy (Bialkowski, 1996) relies on detecting small variations of refractive index and may be used for intracavity sensing (Dudzik et al.,

2021). Photoacoustic spectroscopy consisting in replacing the optical detector by an acoustic detector is a solution to these problems that has been extensively studied in the GSMA (Grossel, Zéninari, Joly, et al., 2007; Grossel, Zéninari, Parvitte, Joly, and Courtois, 2007; Zéninari et al., 2010).

Another way to avoid these problems is called the EVIS technique (External - cavity QCL Voltage Intracavity Sensing) (Phillips and Taubman, 2012; Phillips, Taubman, and Kriesel, 2015). It is based on the self-mixing effect where the source radiation is sent back into the optical resonator, changing the source properties. This effect was demonstrated in semiconductor laser in 1995 (Rochford and Rose, 1995) and in QCL in 2002 (Hofstetter, Beck, and Faist, 2002). For QCL this feedback induces electrical variations. By applying this property to a QCL mounted in an intracavity gas sensing setup, it can be observed that light intensity variations due to gas absorption lead to electrical variations. Thus, the gas spectrum can be retrieved by recording electrical variations of the QCL. To our knowledge, one group has successfully used this technique successfully in the Mid-IR (Phillips and Taubman, 2012; Phillips, Taubman, and Kriesel, 2015), but the remaining problem is the spectral resolution limited by the QCL mode-hops. Indeed, residual reflectivity of the QCL front face is high enough to create a Fabry-Pérot cavity. Different techniques exist to perform mode-hop free spectroscopy with an external cavity setup and are well developed (Tsai and Wysocki, 2012; Wysocki et al., 2005; Liu and Littman, 1981; Gong et al., 2014). Nevertheless, these techniques are strongly limited mechanically by the range of the translation and electrically by the Joule heating, which allows mode-hop free over a very limited spectral range. Usually it is performed over small spectral ranges and the whole spectrum is retrieved by associating them.

In this article we report the development of an EVIS system to detect methane under laboratory conditions. We propose different acquisition and processing methods in order to compensate the mode-hops influence without any mode-hop free technique. The detection is realized over the entire emission spectral range of the laser source. First results are presented and discussed.

2 Experimental setup

The experimental setup consists in a QCL mounted in an external cavity in Littman-Meltcaf configuration and is presented in figure 1. The wavelength tuning is obtained by the association of a fixed grating and a rotating mirror. The external cavity is 38 cm long to allow the insertion of a 20 cm gas cell.

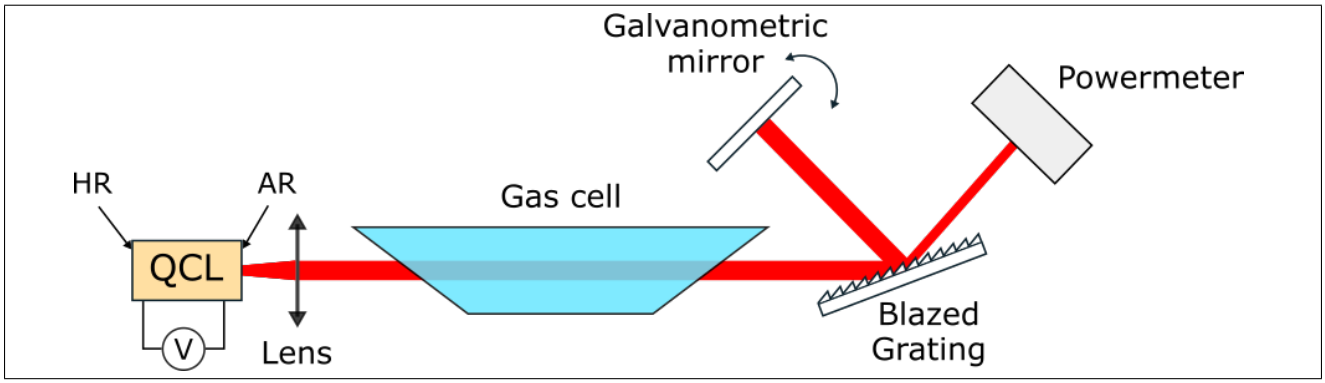

Figure 1: Diagram of the EVIS setup

The QCL chip from mirSense is mounted in a Laboratory Laser Housing from Alpes Lasers with home-made modifications to suit the chip size. The rear chip facet has a high-reflective coating and the front facet has an anti-reflective coating with a R=0.5% residual reflectivity.

A 390037-F Thorlabs lens mounted on a XYZ translation stage is used to collimate the strongly divergent beam from the QCL front facet. The 51029 grating from HORIBA is blazed for 8 µm at 36.52° with 150 grooves/mm. The measured mean efficiency is 70% in the 1st order. In order to adjust the grating orientation, it is mounted on, from top to bottom : a piezoelectric translation, a Thorlabs GNL18/M goniometric rotation and a Thorlabs CR1/M-Z7 closed-loop mechanical rotation stage. The 6220H rotating mirror from Cambridge Technology is gold coated and mounted on a closed-loop gal-

vanometer system. This way, fast running up to 500 Hz and high repeatability with a precision of 8 µrad (4.6×10^{-4} degree) are ensured.

The external cavity laser is operated in continuous mode. The chip temperature is controlled by a Newport LDT-5980 thermal controller with a PT100 probe and a peltier. The temperature is stabilized at 20 °C. The QCL current is controlled by an ILX Lightwave LDX-3232 stabilized current supply. Figure 2 presents the voltage and power characteristics of the EC-QCL when emitting at 1310 cm^{-1}. The EC-QCL threshold is 620 mA and the output power reach almost 11 mW at 840 mA. For higher currents the output power drops due to the Joule heating.

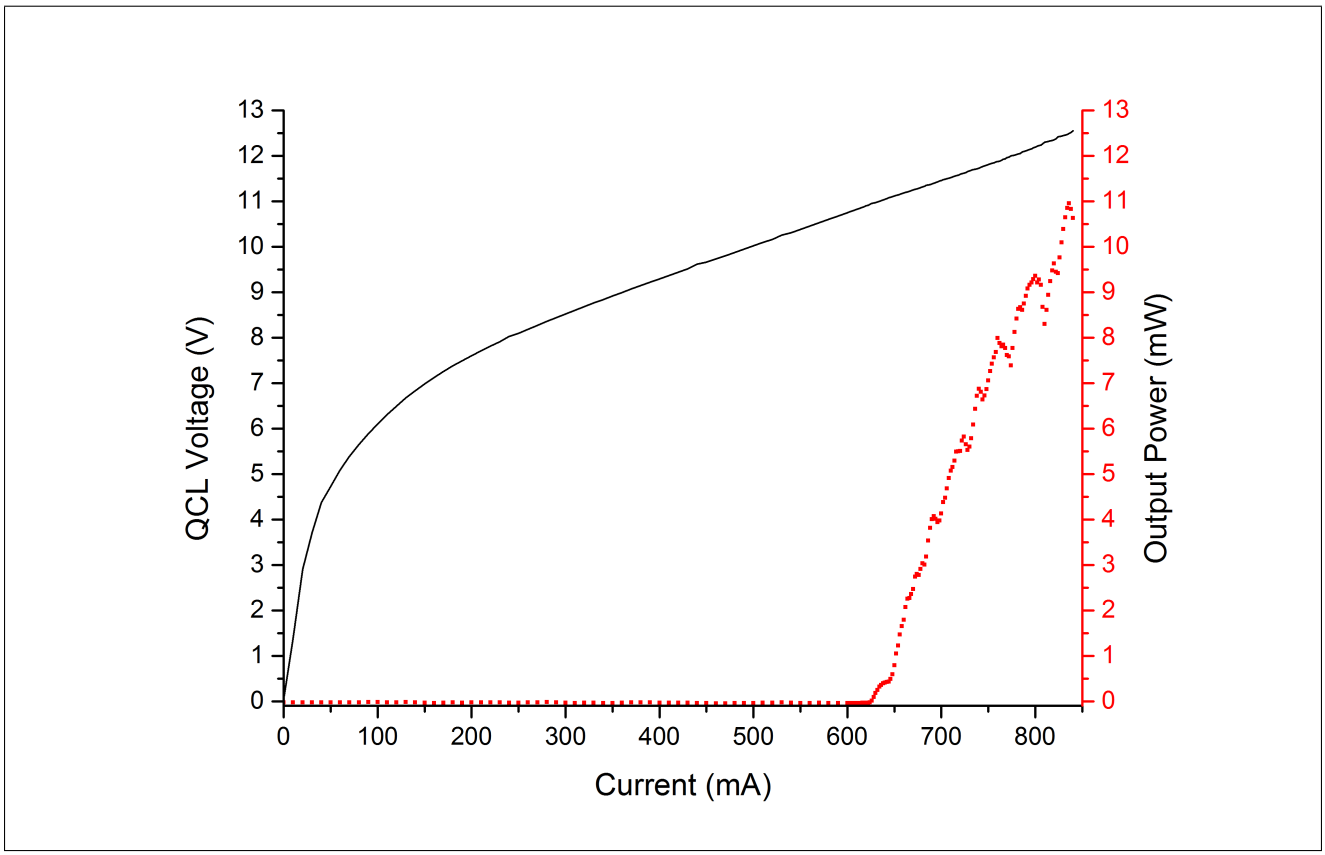

Figure 2: Voltage-Current characteristic of the QCL (black line) and Power-Current characteristic of the external cavity (red dots). Both curves are recorded with the feedback of the external cavity at 1310 cm^{-1}

For the intracavity spectroscopy measurements, a gas cell is inserted inside the external cavity. To minimize laser absorption, the cell windows are made of and mounted at the Brewster angle. The gas samples were prepared using a Gasmix Aiolos from Alytech with an uncertainty lower than 2% of the targeted concentration.

The entire experiment is controlled by custom LabVIEW programs. A FPGA PXI-7841 card is used for the generation and acquisition of signals with a sample rate up to 200 $kS.s^{-1}$. Because the input voltage of the FPGA card is limited to 10 V, a custom differential circuit is implemented to subtract a fixed voltage. The value of this voltage is equal to the QCL voltage with no feedback from the external cavity. The same circuit is used to apply a multiplication factor of 10 after the subtraction. In this way, the output voltage is equal to 0 V when the cavity is not emitting and voltage variations are amplified to improve observations.

3 Characterization of the EC-QCL using EVIS Data acquisition and post-processing

The EC-QCL presented in previous section was operated at constant temperature and with the laser current set at 795 mA. The EC-QCL was tuned by scanning the mirror. The control voltage for the

mirror was a triangular signal of 4 V amplitude at 5 Hz and the compliance voltage of the QCL was recorded by the FPGA card with an acquisition rate of 160 kS.s^{-1}. A full scan over the cavity spectral range, with no gas in the cell, is presented in figure 3. The conversion from the mirror control voltage to wavenumber is carried out with a calibration of the cavity output beam with a FTIR spectrometer at a 0.015 cm^{-1} precision and the diffraction grating equation. The cavity emits over a 71 cm^{-1} spectral range from 1277 cm^{-1} to 1348 cm^{-1}. Taking these parameters into account, the sampling step is 7×10^{-3} cm^{-1}. Figure 3 shows an approximately parabolic variation of the laser voltage during mirror scanning. Numerous rapid variations of the laser voltage are superimposed onto the parabolic shape. Several variations are shown in the inset. Those variations are evenly spaced 0.5 cm^{-1} apart, which corresponds to the free spectral range of the QCL chip. They correspond to mode-hops due to the 0.5% residual reflectivity of the QCL front facet. The external cavity free spectral range is 1.3×10^{-2} cm^{-1} so the corresponding mode-hops cannot be resolved in this configuration. Some other variations, located around 1310 cm^{-1}, 1320 cm^{-1} and 1342 cm^{-1} correspond to water vapor present in the open parts of the cavity. The water lines are far from being resolved.

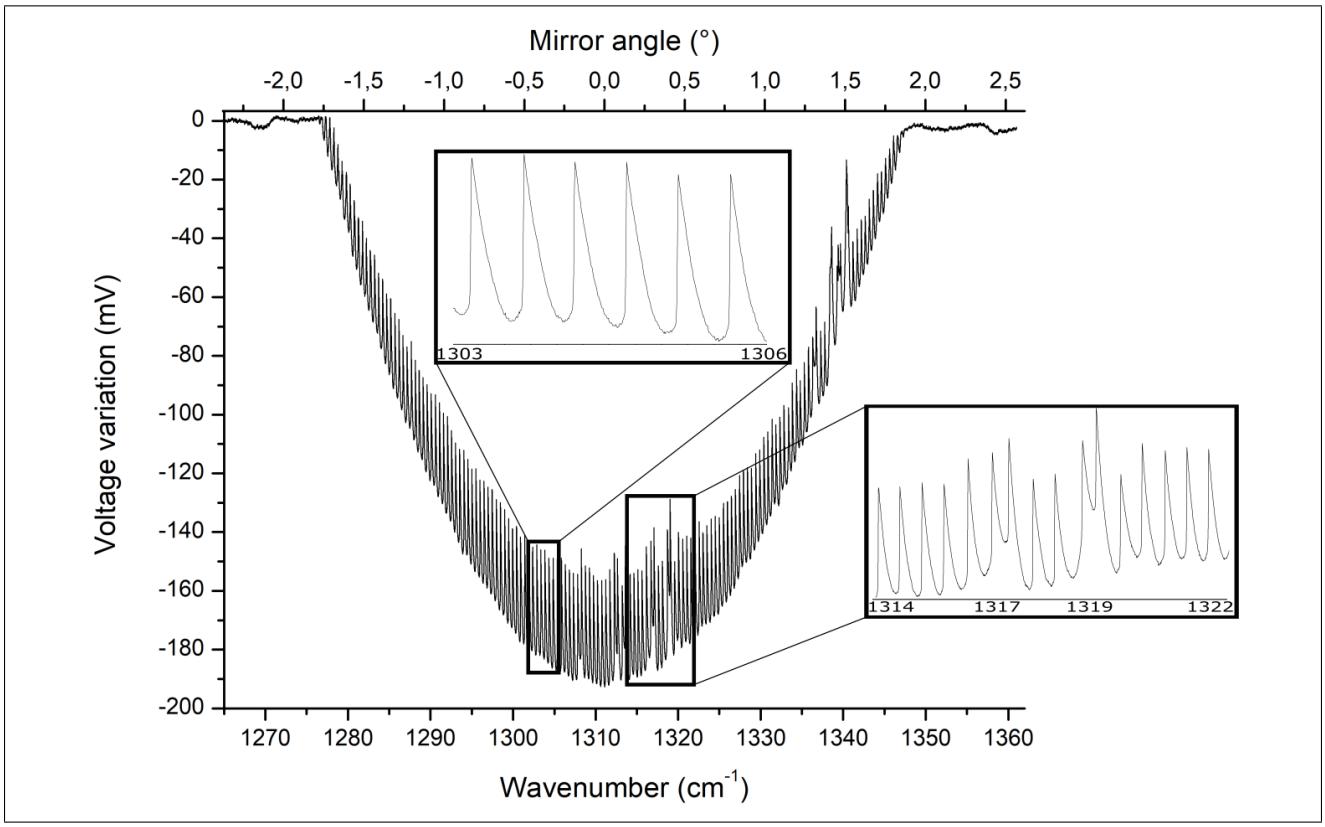

Figure 3: QCL voltage variation for a full scan over the cavity spectral range. Insets show enlargements of parts of the figure. The central one highlights the regular mode-hops pattern. The right one highlights the non-resolved water lines at 1317 cm^{-1} and 1319 cm^{-1}

In order to avoid the influence of the QCL mode-hops, we have developed a data acquisition and processing method based on dual wavelength scanning. The scans are performed simultaneously at different frequencies with triangular signals. The fastest scan is realized with the mirror at 5 Hz over the cavity spectral range. The slowest scan is realized with the QCL current at 0.05 Hz from 795 to 828 mA, this range corresponding to a single QCL mode-hop. For each successive scan of the mirror, the variations associated with the QCL mode-hops are slightly shifted as represented in figure 4. A custom processing program in LabVIEW is used to detect the maximum feedback point between each mode-hop for each mirror scanning. The result of this processing for an empty cell is plotted in figure 5 where the parabolic shape is well retrieved. Some points are clearly found away from the parabolic shape. They are due to the ambient water vapor previously mentioned. Around the top of the water lines the data processing program cannot be applied and wrong points are selected.

In order to obtain gas absorption spectra, two consecutive measurements are performed. First a cavity calibration recording is realized with an empty cell. A second recording is realized with the cell filled by the gas sample. The final step is the voltage difference between the retrieved data points and the

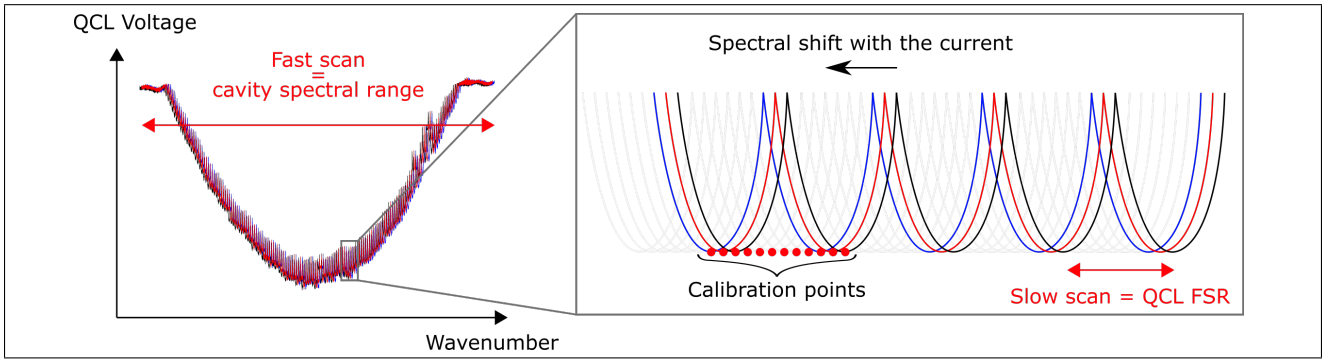

Figure 4: Principle of the dual wavelength scanning

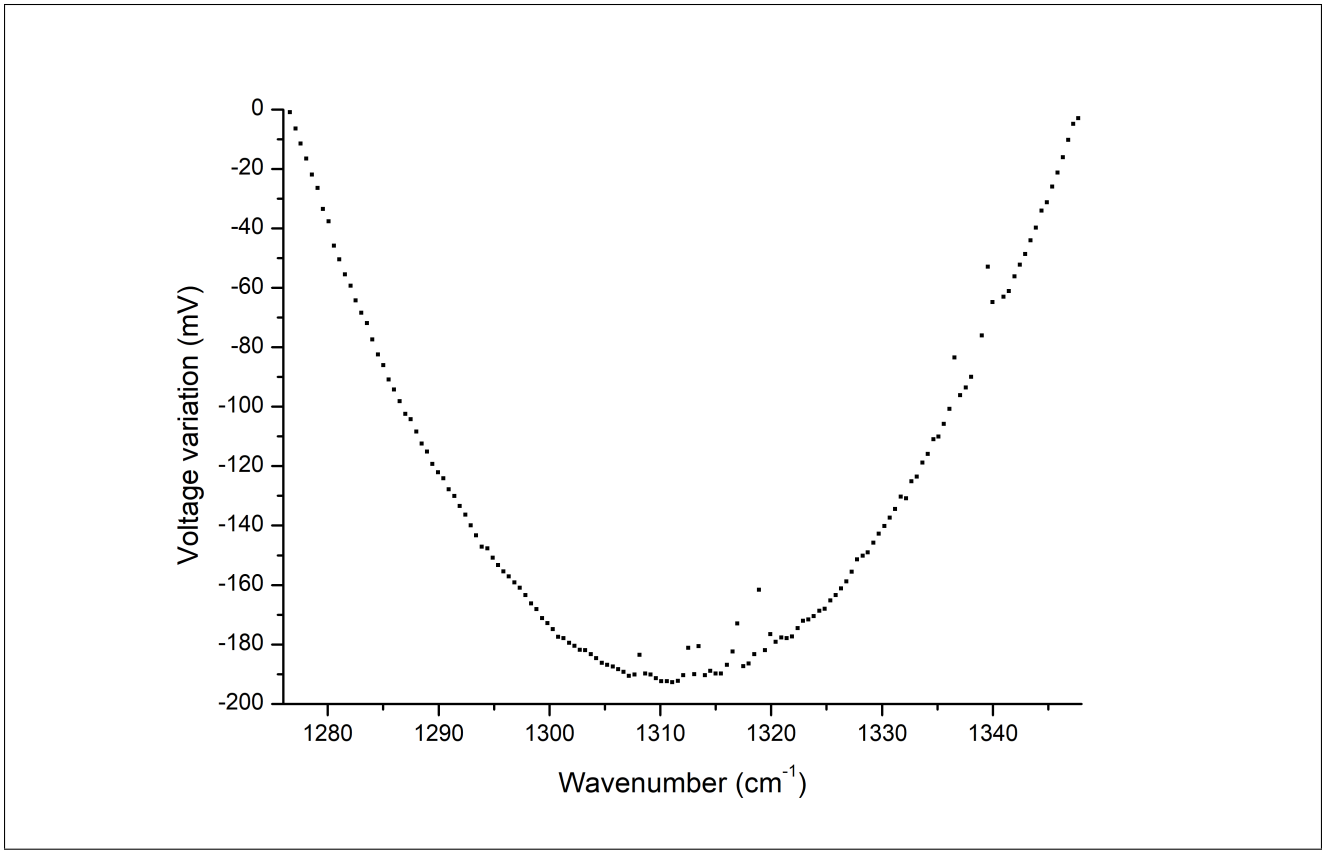

Figure 5: Result of the data processing applied to the data of the figure 3

calibration points. With such a method, the mode-hop influence is compensated in the obtained gas spectrum because each selected point is ensured not to be on a mode-hop.

The measurement uncertainty of the system is studied on consecutive patterns. A voltage noise of 1×10^{-3} V is observed at the end of the acquisition chain. As for the wavenumber uncertainty, it is between 1.3×10^{-2} cm^{-1} and 8×10^{-2} cm^{-1}. This is attributed to a combination of many factors such as the sampling resolution of 7×10^{-3} cm^{-1}, the external cavity mode-hops of 1.3×10^{-2} cm^{-1}, and mainly mechanical instabilities whose effect is enhanced in this long-cavity setup.

4 Preliminary results on CH_4 detection

A spectrum of 0.3% of methane, at atmospheric pressure and ambient temperature, obtained with the described method is presented in figure 6 (top left). The same spectrum without the described method is presented for comparison (bottom left). As expected, the strong influence of the mode-hops is compensated from the spectrum. For comparison, the absorption coefficient calculated with the HITRAN database is presented in the same figure (top right). The water lines visible on the experimental spectrum are due to ambient water vapor in the external cavity open parts. As mentioned previously these lines impact the data processing, therefore the calibration of the system, causing errors in lines amplitude. Thus, in the following we will only focus on the CH_4 spectrum in areas without water lines. For the presented spectrum the resolution is estimated at 2.5×10^{-2} cm^{-1} which is closed to the wavenumber uncertainty as shown in figure 6 (bottom right). For a better comparison, enlargements of parts of the figure 6 are presented in figures 7 (left) and 8 (left). On the right of the same figures is represented the voltage variation versus the absorption coefficient calculated from the HITRAN database. These figures help to highlight the actual limit and uncertainty of the system to retrieve the gas absorption coefficient. This uncertainty is due to the voltage and wavenumber uncertainties mentioned previously. This type of observation was realized in (Phillips and Taubman, 2012) for the slope variation of the voltage-current characteristic and show a similar linear behavior. Further investigation will be conducted to understand these similarities and their limitations. However, from these results we can conclude that our system is currently more suitable for low absorption spectroscopy. The current limit of detection is an absorption coefficient of 5×10^{-3} cm^{-1}. Even if the uncertainty is not negligible, the gas lines are still identifiable over a 69 cm^{-1} spectral range. As one can observe on spectra, some artifacts exist, at 1303.6 cm^{-1} and 1306.2 cm^{-1} for example, which degrade the spectrum quality. Comparing with the theoretical spectrum, this occurs when the linear absorption coefficient is greater than 3×10^{-2} cm^{-1}. We have observed on recordings that a new mode-hop can appear in presence of a strong absorption. This is the consequence of the increase in cavity losses with the increase in gas absorption. An artifact is created when a new mode-hop appears in the area where the maximum feedback was because the mode-hop is retrieved by the data processing program. This explains the dispersion at high absorption observed on figure 8 (right).

5 Conclusion

In this article we performed intracavity gas detection in the mid-infrared region with the EVIS method, consisting in retrieving the gas spectrum through the QCL voltage variations. The main advantages of this technique are the non-use of photodetector and the fact that it works for the full spectral range of the laser source. In addition, it can be easily implemented on an existing cavity without any modification of the optical setup. We retrieved the spectrum of 0.3% of at a 2.5×10^{-2} cm^{-1} resolution over a 69 cm^{-1} spectral range. We have demonstrated the removal of the influence of the QCL mode-hops on the final spectrum with a method based on a dual wavelength scanning. The final resolution is improved by a factor 20, from the 0.5 cm^{-1} QCL free spectral range limitation to 2.5×10^{-2} cm^{-1}. Thanks to this method the external cavity is simple. There is no need for a mode-hop free setup where sensitive mechanical alignments are essential. Moreover, the resolution limit of this method is only limited by the noise and the uncertainty of the acquisition system. The influence of optical cavities inside the setup is compensated. Due to the wavenumber resolution of this experiment the external cavity mode-hops are not resolved. We also used the direct voltage difference in this EVIS setup and compared the obtained results with previous experiments. Similarities were observed between this

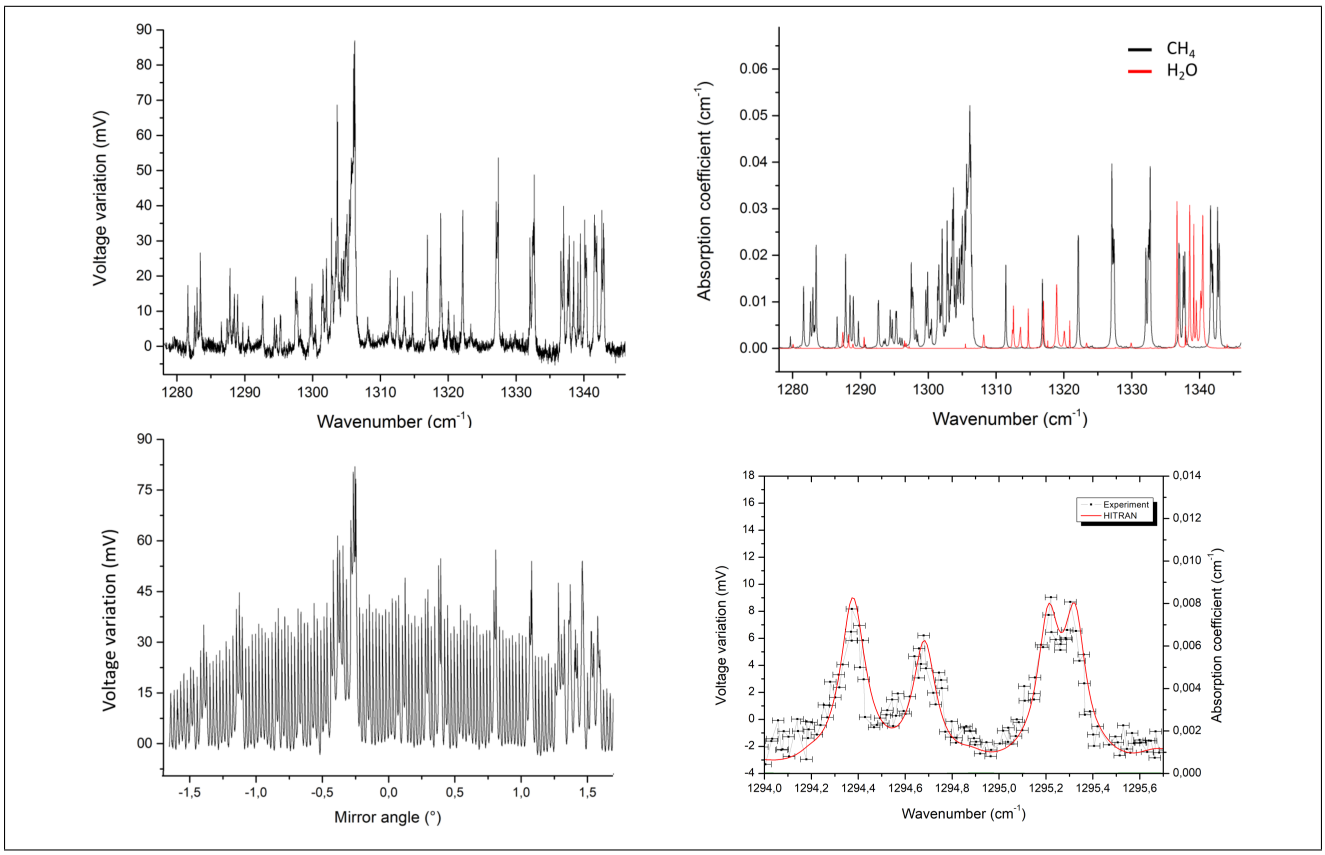

Figure 6: Top Left : Experimental spectra of 0.3 % of CH_4 and ambient H_2O. **Top Right** : Linear absorption coefficient calculated from the HITRAN database. A H_2O concentration of 3 % at ambient temperature and atmospheric pressure is estimated for the calculation. **Down Left** : Experimental spectra of 0.3 % of CH_4 and ambient H_2O without the compensation of the QCL mode-hops. **Down Right** : Enlargment of the top left figure and comparison with the calculated absorption coefficient from the top right figure. The value for error bars is $2.5 \times 10^{-2}\,\mathrm{cm}^{-1}$

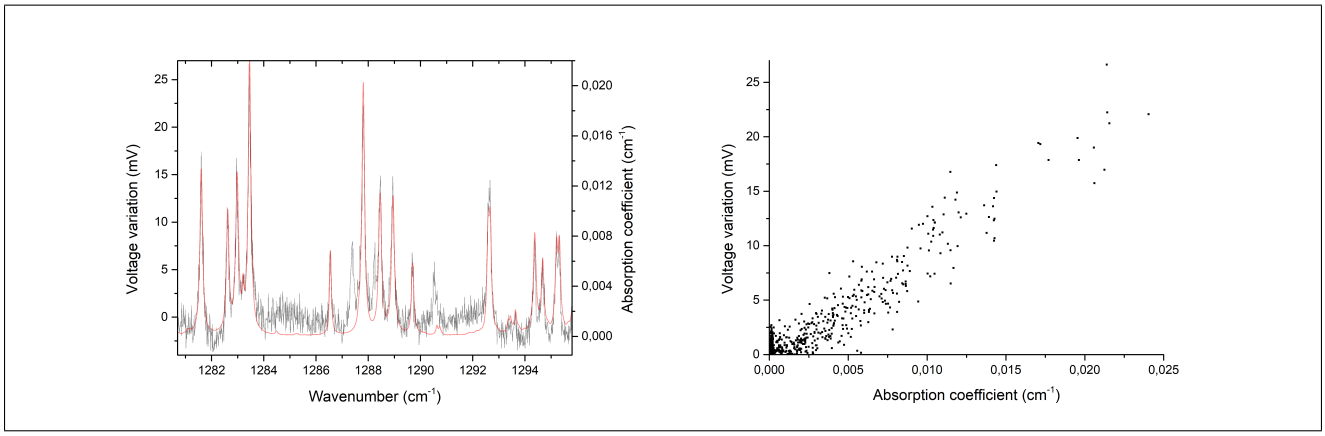

Figure 7: Left : Enlargement of figure 6} (top left). Black line is the experimental spectrum and red line is the absorption coefficient calculated from the HITRAN database. **Right** : Experimental spectrum versus the calculated absorption coefficient for the enlargement.

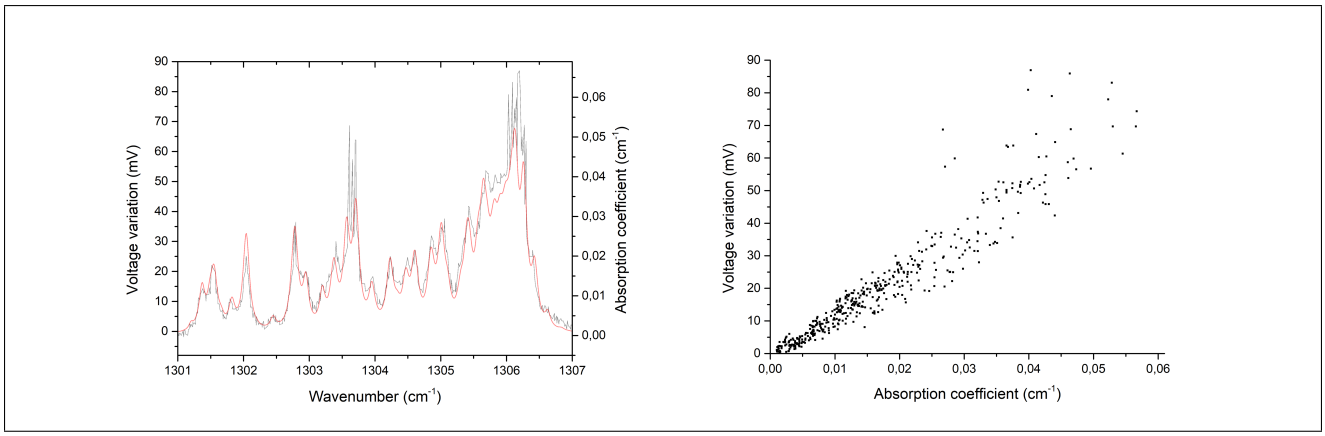

Figure 8: Left : Another enlargement of figure 6} (top left). Black line is the experimental spectrum and red line is the absorption coefficient calculated from the HITRAN database. **Right** : Experimental spectrum versus the calculated absorption coefficient for the enlargement

direct voltage difference and the current-voltage slopes difference, which are promising results. Further studies will be performed to explore the similarities and divergences of these two methods. In order to deal with these results in depth, multiple upgrades are necessary. The problem of the ambient water vapor can be solved by placing the cavity inside a hermetic tank filled with a neutral gas or emptied. The external cavity should also be shortened to reduce the impact of mechanical instabilities.

6 Acknowledgements

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