Appl. Phys. B (2017) 123:145
DOI 10.1007/s00340-017-6716-9

Applied Physics B
Lasers and Optics

# Multi-gas sensing with quantum cascade laser array in the mid-infrared region

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Received: 31 January 2017 / Accepted: 29 March 2017 / Published online: 18 April 2017


**Abstract** Wide tunable lasers sources are useful for spectroscopy of complex molecules that have broad absorption spectra and for multiple sensing of smaller molecules. A region of interest is the mid-infrared region, where many species have strong ro-vibrational modes. In this paper a novel broad tunable source composed of a QCL DFB array and an arrayed waveguide grating (also called multiplexer) was used to perform multi-species spectroscopy (CO, $C_2H_2$, $CO_2$). The array and the multiplexer are associated in a way to obtain a prototype that is non-sensitive to mechanical vibrations. A 2190–2220 $cm^{-1}$ spectral range is covered by the chip. The arrayed waveguide grating combines beams to have a single output. A multi-pass White cell was used to demonstrate the efficiency of the multiplexer.

## 1 Introduction

Quantum cascade lasers (QCLs) are today the only semiconductor laser sources operating over the mid-infra-red (MIR) wavelength range (3–12 μm) at room temperature in either pulsed or continuous wave (CW) mode. Among the available laser technologies such as optical parametric oscillator (OPO) or difference frequency generation (DFG), QCLs have a potential for extreme compactness, high lifetime, high reliability and low cost. Fundamental MIR absorption lines are stronger than the harmonic ones in the near infra-red (NIR). Thanks to the combination of QCL in the MIR and high sensitivity optical methods, MIR absorption spectroscopy can meet the stringent sensitivity and selectivity requirements for air pollutants analysis and green house gases survey. However single frequency operation of the laser is required for standard spectroscopic applications. This is obtained with the development of single mode distributed feedback (DFB) QCLs. Unfortunately the maximum tunning range achieved by changing the laser injection current is only 3–4 $cm^{-1}$ which makes difficult to scan and to allow simultaneous detection of several gas species with a single DFB QCL.

One way to take advantage of the broadband gain of QCLs (from 100 to 200 $cm^{-1}$) is the use of an external cavity (EC) configuration to obtain a single mode operation at any wavelength within the laser gain profile. EC-QCLs are widely tunable but remain cumbersome and complex to build despite recent advances [1–4]. They require high quality antireflection coatings, well-aligned external optical components including a grating for tuning, and piezoelectric controllers. The other way to take advantage of the broadband gain of QCLs has been initially proposed by Lee et al. [5]. Their device was based on an array of DFB QCLs with closely spaced emission wavelengths spanning the gain bandwidth of the QCL material, fabricated monolithically on the same chip and driven individually by a microelectronic controller. The separate beams emerging from the individual lasers in the array were not combined into a single beam. This makes their device not compatible with the actual spectroscopic systems. Several solutions are available to combine separate beams, using either discrete optical elements (lens and grating) [6] or integrated solutions.

This article is part of the topical collection "Field Laser Applications in Industry and Research" guest edited by Francesco D'Amato, Erik Kerstel, and Alan Fried.

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Although high quality results have been obtained using discrete optical elements [7], the overall setup remains cumbersone and cannot be easily miniaturized.

On the contrary, integrated solutions are much more compact and can be produced on an industrial scale i.e. for mass deployment. Different kinds of integrated solutions to combine beams have recently been developed for mid-infrared emission range [8–10]. These integrated devices can be associated to standard DFB QCLs or to sampled grating DFB (SGDFB) lasers [11, 12]. In this paper an arrayed waveguide grating (AWG or multiplexer) directly fixed on the chip will be used as a solution to combine beams [10]. The chip used emits at 4.5 µm, from 2190 to 2220 $cm^{-1}$. CO, $C_2H_2$ and $CO_2$ have spectral lines over this spectral range. Test of the device was made on CO, then spectrum of an industrial cylinder of $C_2H_2$ for torch was performed. Finally $CO_2$ spectrum was recorded using a White cell in order to test the device with a setup sensitive to alignment. Spectral tuning via temperature tuning was chosen over current tuning in order to improve spectral range despite long-time recording.

## 2 Experimental setup

The experimental setup is based on standard laser absorption spectroscopy experiment (Fig. 1).

A room temperature QCL array from MirSense is mounted onto a homemade water cooling with single stage peltier element. It is formed of 9 DFB QCL with different DFB grating in order to have different wavenumbers. Each laser covers an approximately 3 $cm^{-1}$ wide spectral range by tuning the temperature from 10 to 30 °C. Each spectral range overlaps other ones to obtain a wide continuous spectral range. However each laser has its own output beam so it yields a spatially shift of beams which is a drawback to perform easily spectroscopy. To spatially combine beams, a SiGe arrayed waveguide grating (AWG) from CEA LETI is used. A complete description of the design and performances of the AWG can be found in [10]. The AWG used for this work has a multi mode interference (MMI) couplers at the input of the AWG and has no taper. As mentioned in [10], a spectral shift of 10.68 $cm^{-1}$ of the well guided wavelength is observed between the input and the output of the AWG. This shift was taken into account for the choice of the DFB QCL Array. The AWG is properly aligned with its corresponding laser of the array with the help of an infrared camera and glued on the chip. Nine lasers are combined in a single 3 µm × 3.3 µm output. This output beam is collimated by a 15 mm focal length germanium lens (1 in. diameter) which allows laser recording with a Fourier transform infrared spectrometer (FTIR) (Fig. 2). This spectrometer is a continuous FTIR spectrometer which does not offer the possibility to synchronize data acquisition with a laser pulse. Nevertheless as shown in Fig. 2 all lasers are equally spaced by 3 $cm^{-1}$ at a given temperature. Two gaps at 2202 and 2208 $cm^{-1}$ exist because of two damaged lasers.

Lasers are supplied by a QCL pulser switching unit (LDD100 from Alpes Lasers). In Fig. 3 is showed the laser 1 output power evolution with the voltage command. Due to weak output power a recording of a laser pulse is done by an oscilloscope for each voltage command, then laser power is determined by calculating the area pulse. A reference point is measured at the maximum voltage used (22 µW at 20 V) with a powermeter (model S401C, Thorlabs) to convert area value into watts.

A 51 cm long gas cell with 38 mm diameter $BaF_2$ windows is used in this experiment. Light is detected by a VIGO detector (model PCI-3TE-10.6) just after the cell.

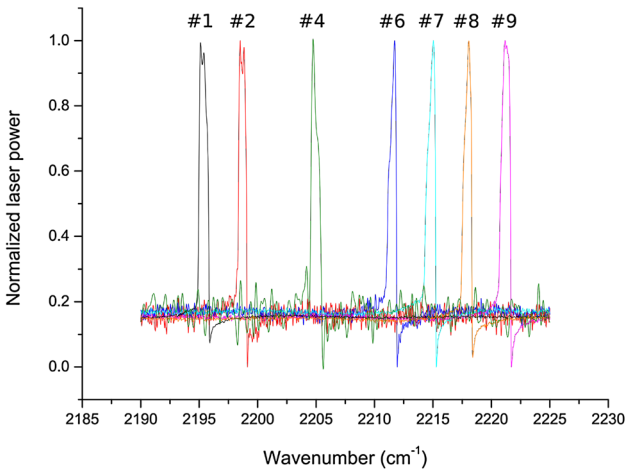


**Fig. 2** Lasers spectra at 20 °C recorded with a continuous FTIR spectrometer. Laser pulses are 50 ns long with a 33.3 kHz repetition rate. Laser number is indicated above laser line

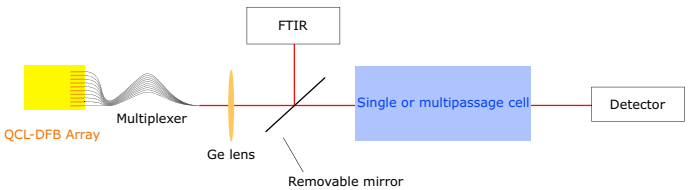


**Fig. 1** Scheme of the experimental setup based on DFB QCL array with a multiplexer

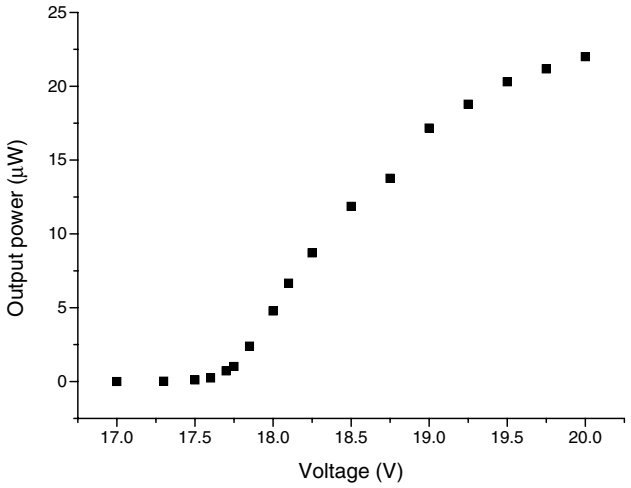


**Fig. 3** Laser 1 output power evolution with the voltage command at 20 °C. Laser pulse is 400 ns long to maximize light power

A boxcar averager (Princeton Applied Research 4121B) with a 1 ns acquisition window is used to limit the effect of spectral broadening due to laser frequency chirping and to accumulate several pulses in order to increase the signal to noise ratio. Note that a setup with two detectors and a beam splitter was not possible because only one boxcar averager was available. The evolution of signal variance versus the number of acquired pulses has been studied for several lasers. This evolution is plotted in Fig. 4 for the laser emitting at 2218 $cm^{-1}$. The slope shows the effect of white noise. The effect of pulse accumulation is more efficient for approximately 100 accumulated pulses. However the variance is still decreasing until 10,000 accumulated pulses and shows no signs of external influence therefore the moving-average was done over 10,000 pulses. These pulses are 400 ns long with a 33.3 kHz repetition rate and 10 µW average power. This last value corresponds to the output power with multiplexer as it cannot be removed.

The boxcar output signal is recorded simultaneously with the chip temperature controlled by a temperature controller (Thorlabs TEC 2000). Recordings are done with constant current operating mode lasers thus wavenumber tunability is obtained by a temperature ramp. Relation between DFB laser wavenumber and its temperature is linear so that the wavenumber can be directly deduced from the recorded temperature. In order to check the wavenumber axis of the spectrum, laser wavenumber is recorded at the begining and at the end of the temperature ramp by the FTIR spectrometer. Signals from boxcar and temperature controller are filtered to reduce noise over lasers intensity and lasers wavenumber. A 10 Hz low-pass filter is applied to intensity signals and a moving-average over 120 samples is performed by the oscilloscope on the temperature signal.

The setup was tested with CO gas at a known concentration and with an unknown gas mixture for industrial application (torch) as presented in the result section. However in order to demonstrate the ease of use of our device and the capabilities of the multiplexer, a more sensitive setup is needed thus a White multi-pass cell is used.

## 3 Results

As explained in the previous section a single-detector setup is used thus two successive measurements are performed with each laser to obtain the transmission spectrum. First measurement is done with an empty cell at less than $2 \times 10^{-2}$ Torr pressure to obtain laser baseline. Since our device is devoted to atmospheric gas detection/gas detection in ambient conditions, the

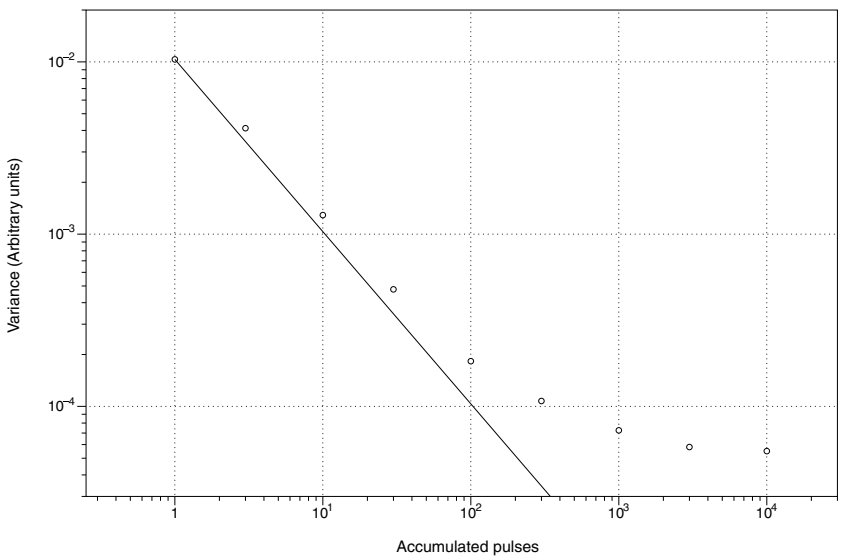


**Fig. 4** Variance evolution with the number of accumulated pulses for the laser at 2218 $cm^{-1}$. Evolution until 100 accumulated pulses is almost like white noise (*black line*) then it derives due to others parameters but still decreases until 10,000 accumulated pulses

second measurement is done with the gas of interest at atmospheric pressure. Finally transmission spectrum is retrieved through the ratio of these two measurements. As wavenumber temperature tuning is chosen in order to improve spectral range a 28 $cm^{-1}$ spectral range is covered by multiplexed lasers but a 90 s recording time is needed for each laser.

To begin with a simple spectrum gas, carbon monoxide which has strong well-separated lines in lasers wavenumbers region was chosen. CO spectrum at 2800 ppm in nitrogen recorded with the multiplexed QCL-DFB array is presented in Fig. 5.

During the temperature ramp, laser power is changing and yields a non-constant signal to noise ratio. Therefore a wavenumber-dependant evolution of this ratio varying from 7 to 25 is observed on lasers spectra. A least square fit using spectroscopic parameters from HITRAN 2012 [13] database is applied to the experimental spectrum in order to retrieve CO concentration (Fig. 5). The spectrum obtained in this conditions cannot be perfectly fitted and a 2400 ppm concentration is retrieved instead of 2800 ppm. This difference can be explained by the offset shift of the boxcar output signal which cannot be monitored during the recordings. Moreover as the fitted lines are close to saturation the fitting is harder. However the main conclusion of this part is that despite the small difference between the experimental value and the fitted one, a whole agreement is obtained thus demonstrating the possibilities to detect CO with this laser source.

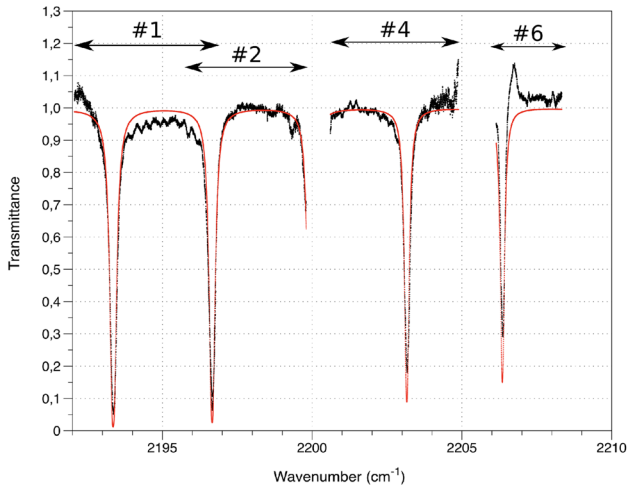


**Fig. 5** CO spectrum recorded with the multiplexed array (*black*) and fit based on HITRAN 2012 data (*red*). CO gas concentration is 2800 ppm at atmospheric pressure. Path length is 51 cm. Spectral coverage for each laser is symbolized above spectrum by a two-headed line with the laser identification number. If lasers 3 and 5 were still running two holes in the spectrum would have been filled. Etalons on laser spectra are due to the lens. Indeed the distance between the multiplexer output and the lens is 1.5 cm which leads to a free spectral range of 0.33 $cm^{-1}$

With the purpose of atmospheric gas detection it is important to demonstrate multi-gas sensing performances of our source. To reach that objective an unknown flame gas mixture mainly composed of dissolved acetylene and impurities (<‰) typically used for industrial application like torch is used. Spectrum obtained with our device (Fig. 6) is fitted with the same method as before.

Because following spectra have more lines than CO, overlaps between lasers are removed to allow better readability of figures. First one can remark the very good agreement between experimental spectra and calculated ones. A composition of almost pure $C_2H_2$ with 400 ppm of CO is retrieved. This result seems logical taking account of the standard composition of this type of mixture. To confirm that CO in the experimental spectrum was not a contamination from previous experiments a mixture spectrum was recorded with the FTIR spectrometer. Result is presented in Fig. 7. FTIR spectrum shows the same absorption due to almost pure $C_2H_2$ and some CO lines corresponding to 400 ppm of CO.

Finally an other property of multiplexed arrays, in addition to multi-gas detection, is their ease of use to perform spectroscopy thanks to the single output. To show that property a setup sensitive to alignment is used. A White multi-pass cell composed of three 50 cm curvature radius gold mirrors in a 5.1 m interaction length configuration is used. Lines absorptions are too strong for both CO and $C_2H_2$ in multi-pass configuration and lead to absorption saturation. On the opposite $CO_2$ has lines that are not saturated with 5.1 m interaction length, moreover it also has an atmospheric interest. Once again one can remark the very

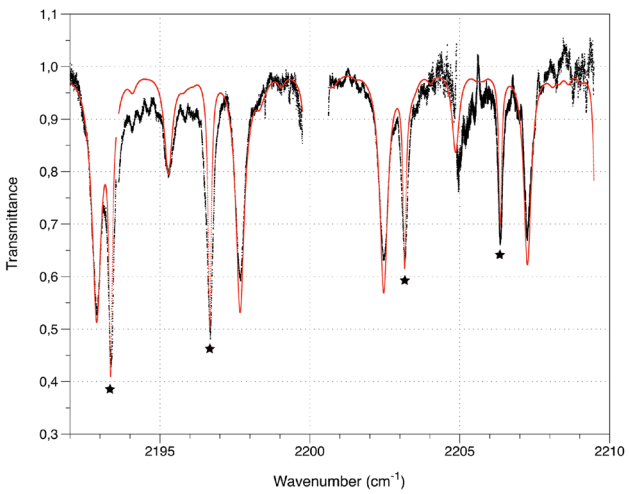


**Fig. 6** Flame gas mixture spectrum recorded with the multiplexed array (*black*) and fit based on HITRAN 2012 database (*red*). Gas is at atmospheric pressure and the interaction length is 51 cm. CO lines are identified by *small black stars* under them. Other lines are $C_2H_2$ ones

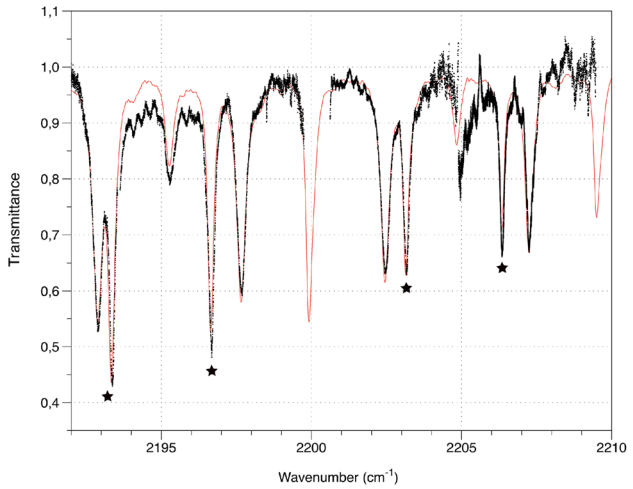


**Fig. 7** Flame gas mixture spectrum recorded with the multiplexed array (*black*) and with the FTIR spectrometer (*red*). Gas is at atmospheric pressure and the interaction length is 51 cm. CO lines are identified by *small black stars* under them. Other lines are $C_2H_2$ ones

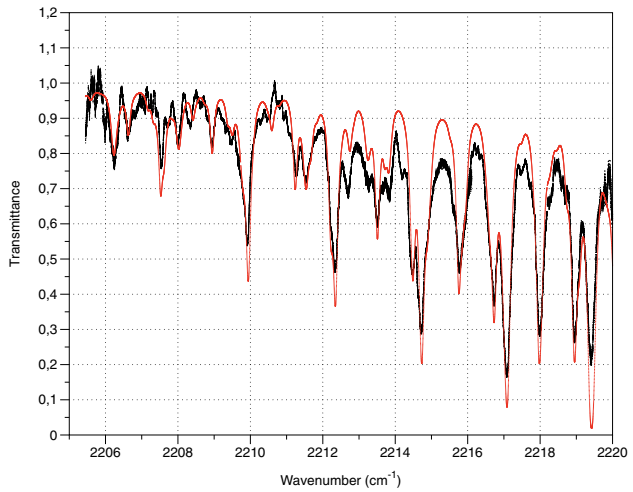


**Fig. 8** $CO_2$ spectrum recorded with the multiplexed array (*black*) and fit based on HITRAN 2012 database (*red*). Gas is at atmospheric pressure and the interaction length is 5.1 m

good agreement between experimental spectra and calculated ones. The fit of the spectrum obtained Fig. 8 yields a ratio equal to the natural abundance with a 1% precision (99% $^{12}CO_2$ and 1% $^{13}CO_2$).

## 4 Conclusion

A multiplexed QCL DFB array was used to perform spectroscopic measurements in the mid-infrared region at atmospheric pressure. Multispecies detection was demonstrated using the source in association with a single path cell. A very good agreement was obtained between the CO and $C_2H_2$ concentrations and the retrieved ones obtained by the fitting of experimental spectra. Moreover the multiplexer added to the array was very useful when a multipass cell must be used for spectroscopy. This has been demonstrated on $CO_2$ and a very good agreement was found between retrieved concentrations and standard ones. Moreover this work demonstrates that this kind of source has also a great potential for measurement of big molecules that do not possess thin absorption lines in the mid-infrared. As a first proof of concept the prototype used in this work shows a lot of possibilities for multi-gas sensing. The next step is the improvement of the covered spectral range, first up to 100 $cm^{-1}$ then up to 200 $cm^{-1}$.

**Acknowledgements** This work was funded by the ANR ECOTECH Project ANR-11-ECOT-004 called MIRIADE (2012–2015). Laurent Bizet also acknowledges Direction Générale de l'Armement and Région Champagne-Ardenne for his PhD funding.

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