

New Quantum Cascade Laser package for Heterodyne Phase Sensitive Dispersion Spectroscopy with High Performance in Free Space

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Abstract: We present a heterodyne phase sensitive dispersion spectroscopy instrument, based on a new QCL package, capable of providing, for the first time, high levels of performance at ambient pressure, thus, enabling free space gas analysis. © 2023 The Authors.

1. Introduction

Molecular scattering spectroscopy, which encompasses such methods as Heterodyne Phase Sensitive Dispersion Spectroscopy (HPSDS) [1-3] or Chirped Laser Dispersion Spectroscopy (CLaDS) [4], has demonstrated in recent years very substantial advantages over traditional spectroscopy methods. In addition, and especially in the case of HPSDS, its similarity in terms of architecture to these traditional systems allows the implementation of hybrid systems, where the advantages of the two measurement approaches, absorption and dispersion, are combined to strengthen the performance of the final system [5].

In the most practical implementation of molecular dispersion spectroscopy systems, the laser, usually a Quantum Cascade Laser (QCL), is modulated at high frequency to generate the multiple tones that are necessary to spectrally interrogate the refractive index change profile in the surroundings of the molecular transitions of the compound of interest. The different refractive indices experienced by each of the tones induce different phase velocities that generate optical phase shifts between the signals. From the subsequent measurement of these phase shifts, parameters such as gas concentration can be straightforwardly retrieved. However, as expected, the performance of molecular dispersion spectroscopy systems is highly dependent on the ratio of the modulation frequency to the linewidth of the molecular resonance. With most of the compounds of interest exhibiting linewidths of several GHz at ambient pressure, and considering that the modulation frequency has to be in the same order of magnitude, it is not surprising that most systems demonstrated to date use multipass cells in which the pressure can be reduced to decrease the linewidth and, therefore, the need to modulate the laser at such high frequencies. For this reason, in virtually all of these instruments, system performance is suboptimal when attempting to analyze gases at ambient pressure. Several instrumental factors limit the modulation frequency nowadays, including the limited availability of fast detectors and the poor suitability of common High Heat Load (HHL) packaging for high-frequency modulation. Fortunately, recent developments are beginning to overcome these technological challenges. On the one hand, high frequency detectors (> 1GHz) have recently become commercially available. However, finding QCLs packages with high modulation capability is not an easy task.

This paper presents a novel QCL package, developed by the company Alpes Lasers, and an instrument that makes use of it to allow, for the first time, the convenient use of high modulation frequencies in HPSDS systems, allowing its operation at ambient pressure and, thus, enabling the analysis of gaseous samples in free space.

2. Description of the QCL package and the instrument

As discussed in detail in references [1-3], an HPSDS system has a simple implementation characterized by the addition of a high frequency RF signal to the QCL bias current. This generates a three-tone optical signal that is used to sweep the targeted spectral line. The change in the profile of the refractive index of the sample (due to the presence of the target gas) results in a phase shift between the optical tones that is translated in a particular variation of the phase of the detected heterodyne signal. From this variation, the gas concentration can be extracted, with the important advantage of having a high linearity between the output signal and the gas concentration.

The main component of the implemented system, whose block diagram is shown in Fig. 1(a), is a QCL in a special packaging which is characterized by an SMA connection that gives direct access to the chip. The laser is a DFB-QCL from Alpes Lasers, with a grating period 1.2 μm , processed as buried heterostructure. The ridge waveguide is 8.5 μm wide and 2.25 mm long. The device operates as single-mode from -10°C to 50°C and is tunable from 1306 to 1314 cm^{-1} . A high reflectivity coating has been deposited on the back-facet. The laser is packaged in a

custom housing, water-cooled and equipped with a thermo-electric cooler. A SMA connector enables to bring a coaxial cable close to the laser contacts and reduce high frequency losses. The laser is collimated with an IR-coated short focal lens. A Bias-T is employed to combine the sawtooth current coming from the current driver with a high-frequency RF signal. The optical emission of the laser is collimated and brought to the area of interest for proper spectral interrogation of the sample. After detection by a Vigo Photonics HgCdTe amplified photodetector with a cutoff frequency of 1.3 GHz, the heterodyne signal, whose phase is to be monitored, is generated. The way to measure this phase that provides the best performance is obviously the use of a lock-in amplifier. However, to allow, with reasonable complexity and cost, the detection of signals in the GHz range, a second oscillator is used, synchronized with the one used to generate the modulation signal and an RF mixer. This allows the frequency of the beat signal to be lowered to the operating range of any lock-in amplifier.

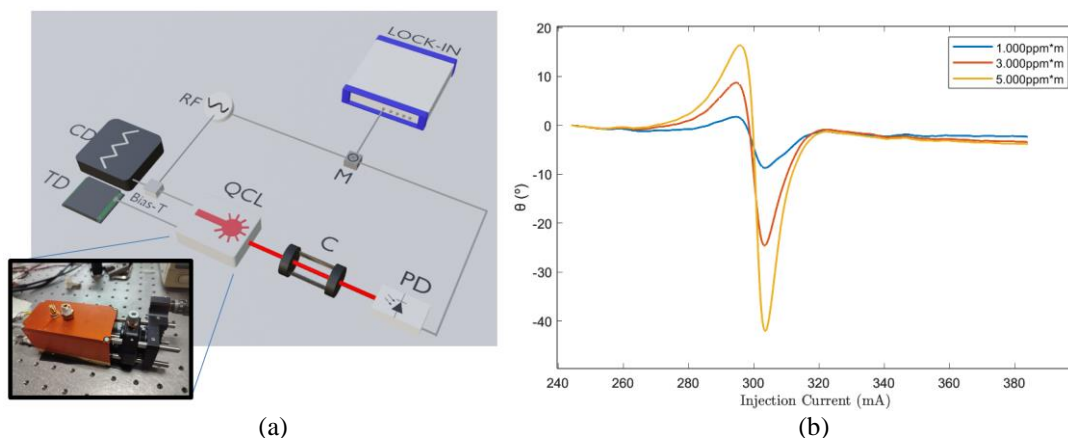


Figure 1. (a) Simplified block diagram of the HPSDS system, TD, Temperature Driver; CD, Current Driver; RF, RF generator; M, Mixer; C, Gas Cell; PD, Photodetector. The inset shows a photograph of the laser. (b) Some results of the calibration process.

3. Experimental validation

The experimental validation consisted in the detection of methane in the vicinity of 1311.5 cm^{-1} . A detailed calibration process has been used to determine the optimum operating point, both in terms of operating current, operating temperature and modulation frequency. For this purpose, a reference cell in which gaseous samples with different methane concentrations can be analyzed was employed. As a representative reference, Fig. 1 (b) shows the output phases for different gas concentrations at ambient pressure, where the linearity of the system can be appreciated even for very high gas concentrations. The laser was operated at a temperature of $10 \text{ }^\circ\text{C}$ and a current of 314 mA; a signal of 1.5 GHz and 10 dB of power was injected into the Bias-T for the direct current modulation of the QCL. Free space measurements can, in the same way, be obtained for different optical paths to characterize the gas concentration in the environment.

4. Conclusion

This article presents the first HPSDS system using the novel LLH packaging developed by Alpes Lasers. This device allows to obtain an instrument that has the possibility to operate with a very high performance at ambient pressure, thus overcoming the limitations of current systems. This further opens up the field of applications of molecular dispersion spectroscopy and will enable its use in new areas and novel applications.

5. References

- [1] P. Martín-Mateos and P. Acedo, Heterodyne phase-sensitive detection for calibration-free molecular dispersion spectroscopy, *Opt. Express* **22**, 209993 (2014).
- [2] P. Martín-Mateos, J. Hayden, P. Acedo, and B. Lendl, Heterodyne Phase-Sensitive Dispersion Spectroscopy in the Mid-Infrared with a Quantum Cascade Laser, *Anal. Chem.* **89**, 5916 (2017).
- [3] W. Ding, L. Sun, L. Yi, and X. Ming, Dual-sideband heterodyne of dispersion spectroscopy based on phase-sensitive detection, *Appl. Opt.* **55**, 8698 (2016)
- [4] G. Wysocki and D. Weidmann, Molecular dispersion spectroscopy for chemical sensing using chirped mid-infrared quantum cascade laser, *Opt. Express* **18**, 26123 (2010).
- [5] A. Genner, P. Martín-Mateos, H. Moser and B. Lendl, A Quantum Cascade Laser-Based Multi-Gas Sensor for Ambient Air Monitoring., *Sensors* **20**, 1850 (2020).

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