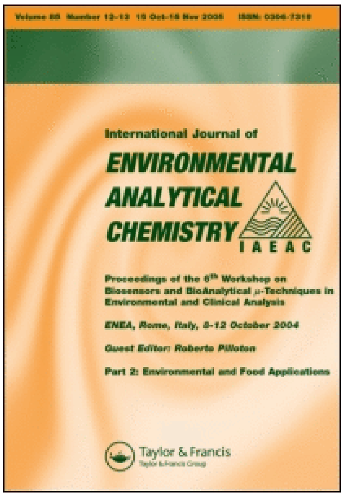


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# ON THE POSSIBILITY OF COMBINING EXTERNAL CAVITY DIODE LASER WITH PHOTOACOUSTIC DETECTOR FOR HIGH SENSITIVITY GAS MONITORING

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An external cavity diode laser is constructed and demonstrated to possess stable, narrow linewidth operation and wide-range tunability, thus being an ideal light source for spectrally resolved detection of absorption lines of small molecules in the gas phase. Moreover, by using this laser in conjunction with a photoacoustic cell, detection limit for optical absorption coefficient about $3.5 \times 10^{-8}$ $cm^{-1}$ was achieved. The performance of the combined system was tested by measuring overtone and combination lines of water vapour, methane and acetylene around 850nm. Because of its cheapness, simplicity and stability, this combined system is a promising candidate for high sensitivity gas detection applications.

*Keywords:* Photoacoustic spectroscopy; diode laser; gas monitoring

## INTRODUCTION

There is an ever-growing interest in high sensitivity gas detection systems. Originally this interest was stimulated by the needs of environmental research[1]. As the sensitivity of gas detection systems has grown to levels below ng/l, the potential for variety of new applications have opened very diverse application areas such as agro-food industry[2] as well as medical research[3]. While the appearance of these new application areas has a positive impact on the development of novel gas analysing devices, it becomes clear that demands on these systems

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diversify to such an extent that it is increasingly difficult to find one universal detector meeting all the different requirements simultaneously.

Photoacoustic (PA) technique, when emerged in the beginning of the '70s was considered for a while to be a very promising candidate for general gas monitoring purposes. Indeed, early papers on PA measurements already reported impressively low detection limits for a large number of gases[4]. The vast majority of these measurements, however, were performed by using gas lasers (mostly CO and $CO_2$ lasers), which rendered most PA systems to remain laboratory devices, hardly suitable for field applications.

It is quite surprising that up to now only few efforts have been made to combine PA detection with room temperature diode lasers[5,6], in spite of the fact that such lasers could be ideal components in a portable system due to their small size, easy operation and long lifetime. Actually, the use of diode lasers in PA systems was hindered by the fact that the PA signal is linearly proportional to the laser power, which is typically three orders of magnitude smaller for diode laser than for CO or $CO_2$ laser. Yet, this inherent loss in sensitivity can be at least partially compensated, because one has the possibility of tuning the wavelength of a diode laser to the peak of an absorption line, while gas lasers are only line tunable and therefore overlap between an absorption line and a laser emission wavelength is only accidental. This, in fact, has been confirmed recently in Ref. 6, where a diode laser based PA detector system was reported with a sensitivity of 8 ng/l for $NH_3$, comparable with that of a $CO_2$ laser based system[7]. The work presented here was therefore stimulated by the results of Ref. 6, and is in line with our previous work on water vapour detection at 1.13 micron[8].

The outline of this article is as follows: first the performance of our home built external cavity diode laser is presented, followed by the description of the PA detector system. Finally the results of the measurements with the combined external cavity diode laser and PA detector system are given.



## EXPERIMENTAL

### The Laser System

Tunability, small size, simplicity, and low cost are the main properties that attracted large interest to use room temperature diode lasers in trace-gas detection applications[9]. Unfortunately conventional, inexpensive Fabry-Perot diode lasers are hardly suitable for spectroscopic measurements because they can be tuned only discontinuously showing "mode hopping" and, besides, they usually have a multimode spectrum which may even change during long term operation ("ageing"-effect). There are diode lasers available with improved spectral properties (DFB or DBR lasers) but their application is far less cost effective. There is an alternative solution of using external cavity diode laser (ECDL) in spectroscopic studies. Such devices have superior spectral properties: narrow linewidth, single mode operation and they are tunable in a wide wavelength range (typically 30-100 nm)[10]. Although an ECDL requires additional elements to be incorporated into the system, their use is still a relatively cost effective solution.

The schematic diagram of the laser system we have built can be seen in Figure 1. This arrangement is known as Littman (or grazing incidence) configuration. The gain medium of the external cavity diode laser was a commercially available, index guided, one facet antireflection (AR) coated diode laser (SDL-AR-5412). The output light from the AR coated facet of the diode laser was collimated with a microscope objective (numerical aperture 0.6) and fell on a diffraction grating (Zeiss, 1800 lines/mm) at near grazing incidence. First order of the diffracted light was reflected back by an aluminium coated mirror to the grating and then to the diode laser. The laser wavelength was tuned by rotating the mirror on a tuning arm. The 0-th order diffraction from the grating served as the output beam with a power of typically 20-30mW and a spot size of 2mm by 4mm. The optical length of the laser cavity was estimated to be about 0.15 m. Temperature and current of the diode laser was controlled by a commercial diode laser controller (SDL Inc.). The whole laser system was built on a 40 cm by 20 cm optical bench without making any special effort to minimise its size. The laser system was optimised mechanically and thermally in order to facilitate its stable and reproducible operation. (Further information on the construction of ECDLs and their application in spectroscopic studies can be found in Ref. 10.)

The linewidth of the laser was measured by using a confocal scanning Fabry-Perot interferometer (TecOptics, FSR 7.5 GHz, Finesse > 200) and was

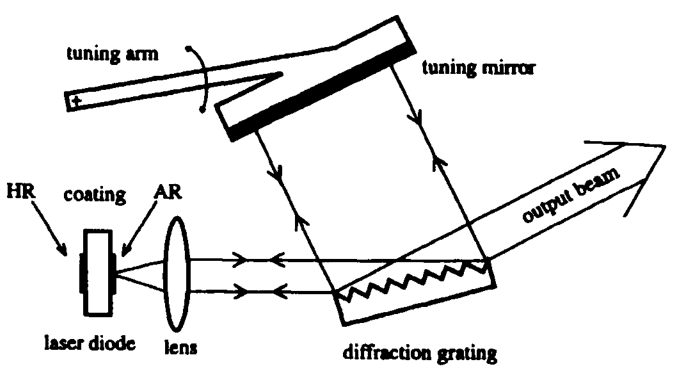


FIGURE 1 Schematic diagram of the external cavity diode laser. (AR: anti reflection coating, HR: high reflection coating)

found to be below the resolution limit of the instrument (about 300 MHz), sufficient for spectrally resolved measurements on absorption lines of small molecules in the gas phase. The same instrument was used for measuring the relative change of the wavelength while tuning the laser. It was found that by properly choosing the length and the pivot point of the tuning arm, continuous, mode-hop free tuning of the laser could be realised in a range exceeding 100 GHz. Linewidth and wavelength was stable even for long time averaged measurements, which indicates the good mechanical stability of the system. The coarse tuning range of the laser was measured with a monochromator and found to be between 835 and 860 nm. Such broad tunability could make possible to perform simultaneous measurements on different components of a gas mixture.

## Photoacoustic Detector System

The operational principle of PA measurements can be understood as follows. The gaseous sample is contained in a closed PA cell which has two openings such that a laser beam can be sent through it. By tuning the wavelength of the laser to an absorption line of the sample, part of the light is absorbed causing local heating of the gas. By modulating the laser intensity the heating also gets periodically modulated which, in turn, leads to periodical changes in the gas pressure i.e. an acoustic wave is generated. The acoustic signal can be measured by using a microphone attached to the PA cell. To increase the measurement sensitivity PA cells are normally designed to be acoustic resonators with light modulation frequency coinciding with one of the resonance frequencies of the cell.

In our PA cell, shown in Figure 2, a 5-mm i.d. tube served as an acoustic resonator. Two acoustic filters (buffer volumes) were attached to its ends to minimise the effect of the background signal caused by possible absorption of the laser



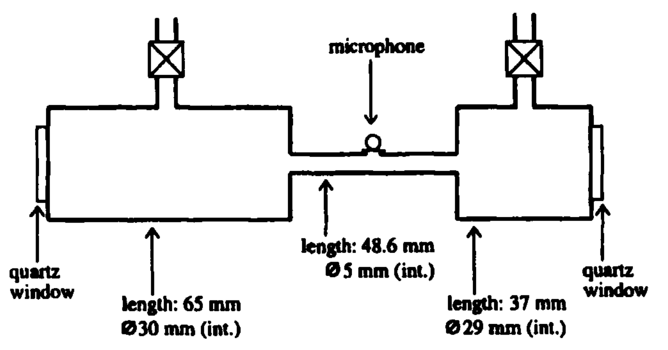


FIGURE 2 Photoacoustic cell for external cavity diode laser based gas monitoring

light on the cell windows. Asymmetrical buffer volumes have been already introduced in diode laser based photoacoustics[6] and proved to suppress both noise and window signal effectively. The small electret microphone (Knowles EA 3029) fixed to the resonator has its highest sensitivity (about 150mV/Pa) in the frequency range from 1 to 4 kHz. The PA cell, which was designed by using a general model developed for the calculation of the frequency response of PA cells[11], has an acoustic resonance around 3 kHz with a quality factor of about 30. The measured frequency response of the PA cell was in good accordance with the prediction of this model. The PA cell has a total length of about 20 cm and a volume of about 100 $cm^3$.

When the PA cell was filled with the specific gas mixture, mostly at atmospheric pressure, its acoustic resonance frequency was measured first. (The resonance frequency can change from measurement to measurement due to changes in temperature, pressure and composition of the gas sample.) The modulation frequency of the laser radiation was then tuned to the peak of the resonance frequency. The intensity of the laser beam was modulated directly by modulating the current of the diode laser. The microphone signal was measured with a commercial lock-in amplifier (EG&G 5110) tuned to the modulation frequency with signal averaging time set to 1 sec.



## RESULTS AND DISCUSSION

In order to test the performance of the combined ECDL-PA system, measurement were made on a mixture of high purity synthetic air pre-mixed with 1% concentration of water vapour. Several absorption lines of water vapour were detected and these lines could be identified by using the HITRAN database[12]. Figure 3 shows the strongest line measured, which was identified as the $7_{1,7} \leftarrow 8_{1,8}$ rotational transition line belonging to the (211-000) vibrational overtone transition at 11962.7 $cm^{-1}$.

One of the advantages of the system presented here is the possibility of performing spectrally resolved measurements of the absorption lines. In this way, it is possible to fit a theoretical line profile to the measured absorption line, yielding enhancement in the sensitivity and the selectivity of the method. Such a fitting was performed by assuming a Lorenzian line shape plus a constant background for one of the measured line (Figure 3) by using a linear curve fitting algorithm. The result of this fitting procedure gave the linewidth (full width at half maximum) of 4.185 GHz, the peak PA signal generated was 24.76 μV after subtracting a constant background of 1.36 μV. This background arises most prob-

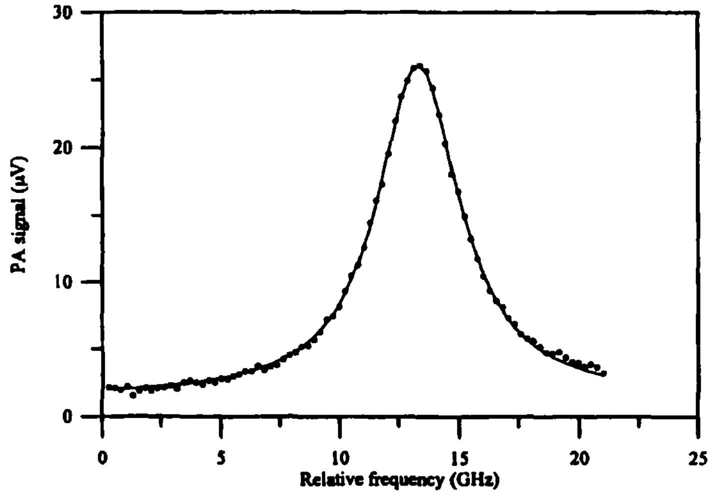


FIGURE 3 Measured water vapour absorption line. Dots represent measured values, solid line corresponds to fitted line profile



ably from absorption of the tail of the beam on the surface of the PA cell. The fitting procedure gave a standard deviation for the peak PA signal: $\sigma$ = 0.18 μV. Due to the well known linear dependence of the PA signal on the optical absorption coefficient[6], $\sigma$ gives not only an information on the accuracy of the measurement for the case of 1% water vapour concentration, but also it can be used to determine the absolute sensitivity limit of the combined ECDL-PA system. Indeed, it can be expected that a water vapour concentration generating a PA signal of 0.18 μV could be detected with a signal-to-noise ratio (S/N) of 1. This corresponds to 70 μg/l detectable concentration of water vapour. From the HITRAN database the measured line has the peak absorption coefficient of 5 x $10^{-6}$ $cm^{-1}$ for the 1% water vapour concentration. Therefore the minimum detectable absorption coefficient can be estimated to be about 3.5 x $10^{-8}$ $cm^{-1}$ with the system presented. The calculated $\sigma$ value coincides fairly well with the measured background acoustic noise level (about 100 nV) in the cell, which was determined while the laser was switched off.

Since in the wavelength range of the present set-up absorption lines of gases with environmental interest are fairly weak, the measurements presented here have to be considered as test measurements on the general performance of the combined ECDL-PA system. It seems to be feasible to increase the sensitivity limit of the combined ECDL-PA detector system into the ng/l concentration range by using diode lasers emitting at longer wavelengths. Especially, the wavelength range of around 1.5 μm could be of significant interest, where several environmentally important molecules have absorption lines that are (5-6) orders of magnitude stronger than the ones measured in this paper[9]. Work is under progress in our laboratory to explore this possibility.

In addition to water vapour, measurements on pure acetylene and methane gases were also performed and a typical $S/N \geq 1000$ was observed. These measured absorption lines have a peak optical absorption coefficient typically $10^{-5}$ $cm^{-1}$ in the 830-860 nm wavelength range[13,14]. In certain cases overlapping between the absorption lines of the different species were found. Therefore in practical gas detection applications on multi component gases the use of signal processing techniques[16], additional to the above described line fitting procedure, might be necessary to maintain the sensitivity and selectivity of the system.

## CONCLUSION



We have demonstrated that external cavity diode laser based photoacoustic detector system is well suited for both gas monitoring applications and molecular spectroscopy. The narrow linewidth and the wide range tunability of the external cavity diode laser make it an excellent light source for spectrally resolved measurements on gas phase molecules. Based on the present measurements of water vapour, acetylene and methane absorption lines in the 835-860 nm region, the minimum detectable optical absorption coefficient was found to be about $3.5 \times 10^{-8} cm^{-1}$ by the combined ECDL-PA system. Such sensitivity can be expected to be sufficient for ng/l level gas detection when using an ECDL at an optimal wavelength range.

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