# Near-IR diode laser-based sensor
# for remote sensing of methane leakage

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A tunable diode laser-based sensor for remote sensing of methane ($CH_4$) leakage at a wavelength of 1.65 μm was developed. The 1.65 μm distributed feedback (DFB) diode laser has a narrow line width and a wide tuning range. It was found that the sensor can detect a 10 $cm^3min^{-1}$ city-gas leak with a sensor output equivalent to the range-integrated concentration over 100 ppm·m.

## 1. Introduction

Tunable diode laser spectroscopy (TDLAS) [1–3] is a useful technique for the remote sensing of trace molecules in the atmosphere. In our laboratory, TDLAS systems for monitoring $CO_2$ have been developed [4, 5].

Methane is a very important gas not only as one of the earth warming gases but also as fuel. Natural gas contains mostly $CH_4$ and is used widely in our daily life as prime energy-source material. From the viewpoint of safety, the detection of $CH_4$ leakage is important. Especially, a wide-area $CH_4$ monitoring system is useful in various places such as chemical plants, gas pipelines, and for preventing disasters in earthquake affected areas. TDLAS can determine the leak position and concentration in a short time. Using the absorption band at 1.65 μm, a compact $CH_4$ TDLAS system can be constructed with the desired performance for the detection of a gas leak within a radius of 200 m.

The absorption band around 3.39 μm is usually employed for the detection of $CH_4$ in the atmosphere (natural concentration level is about 1.6 ppm). The absorption band around 1.65 μm is too small to detect atmospheric $CH_4$. However, the concentration level of $CH_4$ in the atmosphere is dangerous only when it is more than 5%. Therefore, the detection of 1000 ppm order is sufficient to avoid explosion. In such high-concentration monitoring, a very large absorption cross-section is not

required. Though the absorption around 1.65 μm is 2% less than that at 3.39 μm, the sensitivity is sufficient for a gas sensor. The main attractive feature in the case of 1.65 μm source is that a compact tunable near-IR light source such as distributed feedback (DFB) diode laser is readily available. An additional advantage is that fast photodetectors such as InGaAs PIN photodiode are also available in this region. Hence, this system can be used for the wide-area monitoring of $CH_4$ leakage with a high spatial resolution (~15 m).

In the past tunable diode laser (TDL) sensors were based upon the use of multi-mode lead salt diode lasers in the mid- and far-IR spectral regions. These lasers accessed the strong fundamental absorption bands of methane allowing sensitive detection, but at a high cost in the complexity of the instrument. The lasers were coupled to large monochromators to isolate individual lasing modes. Still more complex were to be the instruments used for liquid nitrogen cooling and in cooled detectors. As the near-IR diode laser sources were futher developed and adopted by the telecommunications industry it became possible to utilize the same sources and InGaAs detectors for near-IR overtone absorption spectroscopy. The near-IR diode lasers were single mode devices and allowed the use of fiber optic coupling and transmission technology. The sensor we describe below is based upon this technology.

There are several commercially available, currently-o-the-shelf (COTS) technologies which we have taken advantage of in the development of this sensor. These technologies include telecommunications grade NIR DFB diode lasers, single mode fiber optic components including splitters, collimators and patch cords. The sensor architecture, calibration and resulting sensitivity are discussed below.

## 2. Diode laser-based absorption spectroscopy and wavelength modulation spectroscopy (WMS)

### 2.1. Absorption spectroscopy

The remote $CH_4$ leakage sensor discussed in this publication is based on the quantitative absorption of a near-IR diode laser beam propagating through a gas sample. The transmission is given by the Beer–Lambert relation:

$$T = \frac{I(\nu)}{I(\nu_0)} = \exp\left[-Sg(\nu - \nu_0)\,g(\nu - \nu_0)\,n\,L\right] \tag{1}$$

where $I(\nu)$ is the transmitted intensity at frequency $\nu$ after propagation through a path length, $L$ (cm) is the initial intensity, $S$ (cm/molecule) is the line strength of the transition centered at $\nu_0$, $g(\nu - \nu_0)$ is the line shape function (cm) at pressure $P$ and $n$ is the number density of the absorbing species ($cm^{-3}$). The peak absorption coefficient is given by:

$$\alpha(\nu_0) = Sg(\nu - \nu_0)n. \tag{2}$$

The integrated absorption coefficient is:

$$\alpha = Sn. \tag{3}$$

Since by definition:

$$\int_{-\infty}^{\infty} g(\nu - \nu_0)\,\mathrm{d}\nu = 1. \tag{4}$$

the line shape function $g(\nu - \nu_0)$, depends on the gas temperature and pressure and the absorption line strength depends upon the gas temperature. The water concentration [$CH_4$] in molecules/$cm^3$ is given by:

$$n = \frac{-\int \ln\left[\dfrac{I(\nu)}{I(\nu_0)}\right]\mathrm{d}\nu}{SL}. \tag{5}$$

## 2.2. Wavelength modulation spectroscopy (WMS)

The technology described above is called direct absorption measurements. Conventional direct absorption measurements have to resolve small changes in a large signal. In comparison with direct spectroscopy, the benefits of modulation spectroscopy in TDLAS are two-fold. Firstly, it produces a difference signal which is directly proportional to the species concentration (zero baseline technique) and, secondly, it allows the signal to be detected at a frequency at which the laser noise is significantly reduced. WMS has been used with tunable diode laser sources since the early 1970s. The earliest TDL systems used a modulation frequency in the lower kHz range and second harmonic detection. Today, a 50 kHz modulation with 100 kHz detection is quite usual and, consequently, it is convenient to regard 100 kHz as the limit of conventional wavelength modulated TDLAS. Modulation spectroscopy is characterized by ease with which diode lasers can be modulated.

Wavelength modulation spectroscopy involves modulation of a single mode laser beam at a frequency much less than the width of the absorption line being measured. This modulated beam is then passed through a sample of the gas to be investigated and its intensity is measured using a photodetector. The photodetector output is demodulated using a lock-in amplifier. For relatively weak absorption lines, Eq. (1) can be approximated as:

$$I = I(\nu_0)\left[1 - Sg(\nu - \nu_0)\,nL\right]. \tag{6}$$

The time variation of the modulated laser frequency can be expressed as [4]

$$\nu(t) = \nu_0 + \delta\nu\cos(\omega_m t) \tag{7}$$

where $\omega_m$ is the modulation frequency and $\delta\nu$ is the amplitude of the modulation. The time-dependent part of the intensity can then be expanded in a Fourier series [6]

$$g(\nu_0) + \delta\nu\cos(\omega_m t) = \sum_{N=0}^{\infty} H_N(\nu_0)\cos(N\omega_m t) \tag{8}$$

where $H_N(\nu_0)$ is the $N$-th component of the Fourier series.

After detection by the lock-in amplifier at the $N$-th harmonic of the modulation frequency, the signal is proportional to [7]

$$\text{Signal} = \frac{I_0 L}{N!}\delta\nu^N n S g^N(\nu) \tag{9}$$

where $g^N(\nu)$ is the $N$-th derivative of the line shape function. Due to the atmosphere pressure used in our experiments, the line shape can, to a good approximation, be considered Lorentzian. The $N$-th derivative of a Lorentzian line can be written as [5]

$$g_L^N = \left(\frac{\Delta\nu}{2\pi}\right)\frac{-1^N}{y^{N+1}}\sum_{k=0}^{[N]/2} C_{2k,N}\left(\frac{\Delta\nu}{2}\right)^{2k}(\nu-\nu_0)^{N-2k} \tag{10}$$

where

$$C_{2k,N} = (-1)^k \frac{(N+1)!}{2k+1}\binom{N}{2k}, \tag{11}$$

$$\binom{N}{2k} = \frac{N!}{(2k)!(N-2k)!}. \tag{12}$$

Substituting the $N$-th derivative of a Lorentzian line into the expression for the signal, given by Eq. (9), and evaluating this expression at line center, we find that for odd harmonics, the signal is equal to zero. These zero crossings at line center make odd harmonic signals convenient for use as error signals to stabilize the laser frequency. While this null crossing can be predicted by considering the symmetry alone, the formulation above gives a quick and simple way of estimating the slope at these zero crossings. The slopes are important in determining the precision with which the stabilization can be implemented. Other factors include noise levels in the feedback loop and tuning characteristics of the laser. Most measurements in modulation spectroscopy are performed at $2\omega_m$, and hence the term $2f$ technology.

## 2.3. Sensitivity

For measurements made with the $2f$ technique at frequencies below 100 kHz, low-frequency noise generally determines the detection limit. The minimum detectable absorption is approximated by

$$\alpha_{\min} = \frac{I_{ex}}{I_0}\left(\frac{B}{f}\right)^{1/2} \qquad (13)$$

where $B$ defines the frequency dependence of the laser excess noise and ranges between 0.8 and 1.5, is the magnitude of the laser power fluctuations at 1 Hz in a 1 Hz detection bandwidth: it is approximately proportional to the laser power and depends on the intrinsic noise of the diode laser and the measuring system. A value of $I_{ex} = I_0 = 10^{-4}$ is typical.

The minimum mixing ratio detectable in a given detection bandwidth is given by

$$M_{\min}(ppbv) = \frac{a_{\min} \times 10^9}{\sigma_0 L N_t} \qquad (14)$$

where $\sigma_0$ is the absorption cross-section at the centre of the monitoring line, $L$ – the path length, and $N_t$ – the total molecular density of the gas. For the $CH_4$ line selected, a detecting limit of a few ppm·m is estimated.

## 3. Sensor design

There are two basic spectroscopic parameters which govern the design of any absorption spectrometer when attempting to achieve a minimum number density detection sensitivity. The first is the absorption line strength $S$, at the anticipated measurement temperature. The reported line strength value for the 1.65 μm absorption feature in the HITRAN database is $10^{-20}$ cm per molecule magnitude [8]. The second important parameter is the collisional broadening parameter $\gamma$, for the bath gas of interest.

In the low pressure limit the absorption line shape is characterized by a Gaussian distribution function with the temperature determining the Doppler width of the absorption function. Doppler line shapes tend to be quite narrow with highly peaked absorbance values. As the pressure of the sample gas rises collisional broadening mechanisms begin to change the spectral width of the absorption line shape, adding Lorentzian character. In the limit of low temperature and finite pressure the line shape would be primarily Lorentzian. Lorentzian line shapes tend to be broad, with depressed peak absorbance values. In a typical sensor operation the line shape falls somewhere between Gaussian and Lorentzian, and is thus described by a Voigt line shape which is a convolution of Gaussian and Lorentzian line shape functions.

### 3.1. Diode laser module

The diode laser module included a diode laser, a diode laser controller, a 1×2 fiber optic splitter, a photodetector, and DC power supplies for the laser operation.

The laser is selected according to three criteria: absorption line sufficiently strong to obtain maximum sensitivity; mode characteristics of the laser emissions as clean and narrow; absence of interference from other gases.

The diode laser operated at 1.65 µm and had a spectral line width of approximately 2 MHz. The laser tuned at approximately 0.03 $cm^{-1}mA^{-1}$ and was typically scanned over 0.9 $cm^{-1}$ to fully capture the methane lineshape. The laser injection current and temperature were controlled by ilx LightWave current controller LDX-3100 and temperature controller LDT-5100. The optically isolated fiber coupled output from the diode laser was split by a 70/30 single mode wavelength flattened fused fiber optic splitter. The output from the 30% leg of the fiber splitter was coupled directly to an InGaAs reference photodiode located within the balanced detector. The 70% leg was connected to the fiber optic launch collimator to detect methane in air. After traversing the absorption cell the signal beam was detected by a second, 3 mm in diameter, windowless InGaAs photodiode.

## 3.2. Computer module

The automated $CH_4$ vapor sensor was controlled by a 1G MHz Pentium 3 industrial rack mounted computer. The system was outfitted with a 300 kHz data acquisition (DAQ) board AC6115 and a ADLink PCI-DIO-96 digital I/O (DIO) board. The DAQ board was used to control the laser diode controller and to digitize signals from the balanced detector. The DIO board was used to implement software control of the digital potentiometer, LDX-3100 and LDT-5100. The sensor control software was developed using National Instruments LabWindow/CVI software programming language. The software provided three separate graphical user interface (GUI) screens for sensor operation including a main control and data presentation GUI, an oscilloscope GUI and a digital filter GUI.

## 3.3. Detector circuit

To decrease the effect of laser noise, we adopt difference balance detection scheme. Here we used a detection circuit which can reduce kinds of noises, background signal. The circuit has large dynamic range to meet the requirement of lock-in amplifer (LIA). A block diagram of the detection circuit is shown in Fig. 1. We established the circuit in our laboratory and compared it to New Focus 2011-FC high performance detector, and found that the performance of our home made detector is better than that of the 2011-FC.

## 3.4. Schematic of sensor

The diagram of sensor is shown in Fig. 2. Tuning and modulation of the laser is accomplished by changing injection current and temperature of the diode laser. The function generator provides an output sine wave at a user specified amplitude and frequency, which is used to modulate the laser beam. This sine wave is also used by the LIA as an internal reference signal for synchronous detection of the signal. The modulation frequency was chosen to be 3 kHz based on a noise analysis and the laser modulation characteristics. The amplitude of the modulation was chosen to be at 0.2 Vrms, which was found to provide the best SNR (signal-to-noise ratio). For
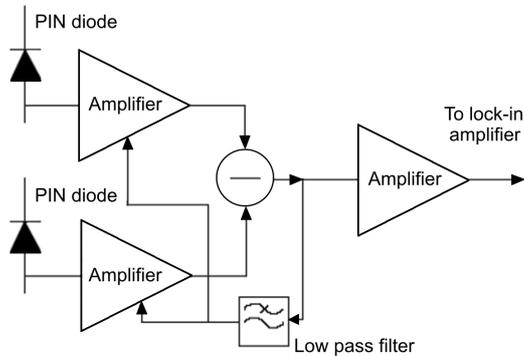
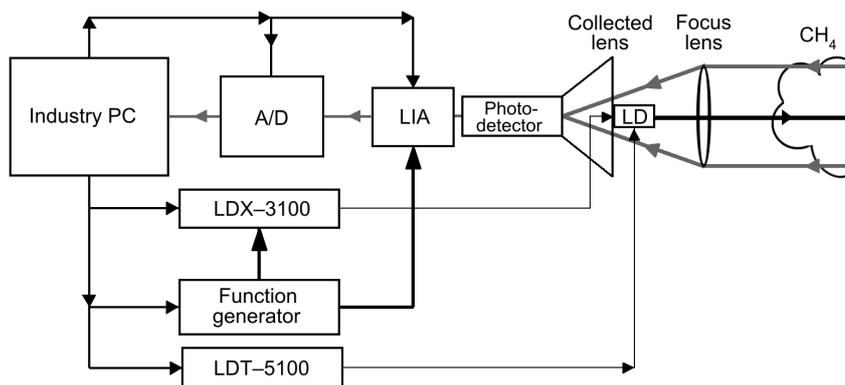Fig. 1. Difference balance detection circuit diagram.



Fig. 2. Schematic of the sensor.

spectroscopic data a voltage ramp is also applied to sweep the laser across the absorption lines being investigated. Part of the laser beam is sampled for wavelength measurement with a Burleigh Wavemeter WA-1500.

## 4. Sensor performance as a gas-leak detector

In order to demonstrate the performance of the sensor as a practical gas-leak detector, we carried out an experiment concerning detection of simulated gas pipeline. Several kinks of reflecting objects were selected as the target and were located within the range of 5 m with the angle of incidence 0°. We prepared a leak point using pure methane in front of the target and set the flow rate at 10 cm$^3$min$^{-1}$. This is the minimum flow rate to be detected in gas-leak detection. We found that the sensor can detect a 10 cm$^3$min$^{-1}$ gas leak with a sensor output equivalent to the range-integrated concentration over 100 ppm·m.

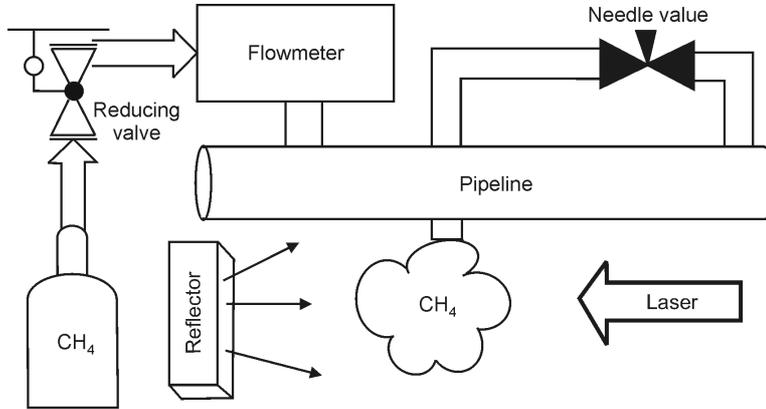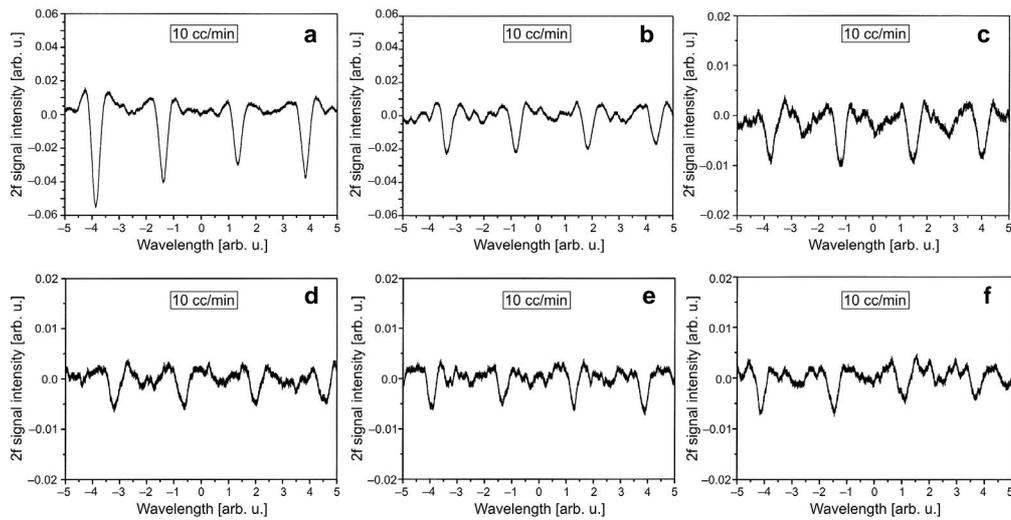Fig. 3. Schematic of pipeline simulation.



Fig. 4. WMS signal of different reflecting objects: brick (**a**), wood board (**b**), bush leaf (**c**), cement plate (**d**), dry sod (**e**), humidity clay (**f**).

We used an aluminum pipe, which is 30 cm-long and whose diameter is 25 mm, to simulate natural gas pipeline. For the main ingredient of natural gas is methane (90%), we chose methane as the main gas to be detected in our labortory. The experimental setup is shown in Fig. 4. The reflecting object we used in Fig. 4 including brick, wood plate, *etc*. We found that most reflecting objects have strong diffuse reflection character and are not sensitive to incidental angle. The typical reflecting objects we used are brick and wood board.

The $2f$ signals of different reflecting objects are shown in Fig. 4. We can conclude from the experiment and Fig. 4 that:

– If the reflectance of the reflector objects varies, the amplitudes and SNR of $2f$ signals are different, too. The higher the reflectance, the higher the SNR. For example, the reflecting signal of brick (Fig. 4**a**) and wood board (Fig. 4**b**) are greater than that of hay (Fig. 4**e**) and clay (Fig. 4**f**).

– The amplitude of $2f$ signal has strong relation to the reflectance of reflector and weak relation with the kinds of reflector.

– From the figure we can see that $2f$ signals of different reflectors with the same leak rate of methane differ from each other, so the reflectance of reflector is a key feature to natural gas pipe leakage detection.

## 5. Conclusions

We have developed a remote methane sensor using a 1.65 μm InGaAsP DFB laser. Wavelength modulation spectroscopy technology is used to get very high sensitivity. The sensor can detect a 10 cm$^3$min$^{-1}$ city-gas leak within a range of several meters. For certain applications, such as methane leaks in coal mines, the sensitivity is more than adequate (*i.e.*, orders of magnitude lower than the explosive limit can be detected). Applications for other gases can evidently be envisaged. If we change the diode laser to a diode laser at other wavelength, for example, at 1.39 μm, we can use the sensor to detect water vapor.

So, in fact, we developed a sensor that can be used to detect $H_2O$ (1.39 μm), $CO_2$ (1.57 μm), $NH_3$ (1.53 μm) and $NO_2$ (0.79 μm), *etc*.

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