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A Compact Remote Methane Sensor using a Tunable Diode Laser

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1 Introduction

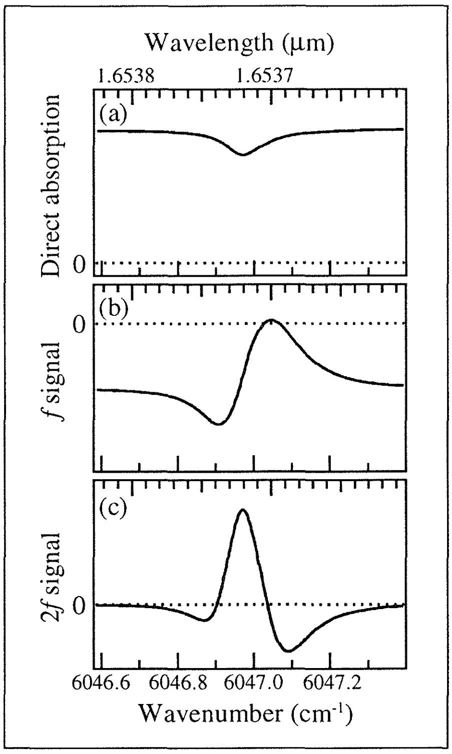
Since methane is an explosive gas and major constituent of natural gas, remote detection of methane is desired for safety in gas facilities and appliances. No conventional gas sensor, however, is capable of remote detection. Only spectroscopic method enables it. Moreover, spectroscopic method has many other advantages such as high sensitivity, fast response, molecular selectivity and so on. In particular, absorption spectroscopy using a tunable diode laser (TDL) has the possibility to realize a compact and low-cost remote methane sensor.

Methane has strong absorption of the ν_3 and ν_4 bands at 3.3 and 7.7 μm, respectively. At present, however, it is still difficult to make a room-temperature TDL of wavelength over 2.2 μm. The strongest absorption of methane below 2.2 μm is the $2\nu_3$ band located in λ = 1.6 - 1.7 μm (Bobin 1972). Furthermore, there are several lines free from the interference of atmospheric gases in this band. Remote detection of methane with one of those lines using an InGaAsP distributed feedback (DFB) laser was reported previously (Uehara and Tai 1992).

In the present work, we have developed a man-portable remote methane sensor using a 1.65-μm InGaAsP DFB laser based on frequency modulation (FM) spectroscopy. The sensor was designed as a long-path absorption lidar using a topographical target with the range of up to 10 m. Because the sensor is very compact and light, an operator can search gas leak by scanning the laser light very easily.

2 Principle of Detection

In FM spectroscopy, laser frequency is modulated at frequency *f* and photodetector output is processed by phase-sensitive detection with reference to fundamental (*f*), second harmonic (2*f*) or higher harmonics. Figures 1 show the direct-absorption (a), the *f* signal (b) and the 2*f* signal (c) of the $2\nu_3$-band R(3) line of methane at the atmospheric pressure observed by temperature tuning of a 1.65-μm InGaAsP DFB laser. In the spectra (b) and (c), the injection current was sinusoidally modulated at *f* = 10 kHz. As shown in



Figures 1. Absorption spectra of the $2\nu_3$-band R(3) line of methane in a 4-cm-long cell at 1 atm observed by temperature tuning of a 1.65-μm InGaAsP DFB laser: direct absorption (a), *f* (fundamental) signal (b) and 2*f* (second-harmonic) signal (c). The modulation frequency *f* in spectra (b) and (c) is 10 kHz.

Figures 1 (b) and (c), the *f* signal suffers a large offset as a result of the amplitude modulation (AM) of the laser. On the other hand, the offset of 2*f* signal is much smaller. Thus the 2*f* signal is more suitable for small absorption detection. Since the 2*f* signal has the maximum value at the center of the absorption line, the modulation-center frequency should be locked at the center of the absorption line to obtain the highest sensitivity. This frequency locking is accomplished by temperature-feedback electronics. The control signal is provided by the *f* signal of the light after passing through a reference methane cell.

We applied the FM spectroscopy to remote detection of methane. The offset of the *f* signal was used to normalize the photodetector output variation caused by the change of taget reflectivity, range, incident angle and so on. The DC component P_{DC}, the *f* amplitude P_f, and the 2*f* amplitude P_{2f} of the received power of the sensor are given by

$$\begin{aligned} P_{DC} &= \frac{A_0 \eta \rho}{R^2} S_{DC} \\ P_f &= \frac{A_0 \eta \rho}{R^2} S_{DC}\, m \\ P_{2f} &= \frac{A_0 \eta \rho}{R^2} S_{DC}\, k \alpha_0 \cdot 2 C_R \end{aligned}$$

where

A_0 : effective area of the colection lens
η : efficiency of the receiver optics
ρ : reflectivity of the target per steradian
R : range to the target
m : AM ratio of the laser
S_{DC} : DC component of the initial power
k : coefficient (depends on FM amplitude)
α_0 : absorption coefficient at the line center
C_R : range - integrated methane concentration.

The range-integrated methane concentration C_R is doubled in P_{2f}, because optical absorption occurs in both ways between the sensor and the target. For Lorentzian lineshape, the explicit expression of the coefficient *k* (Wahlquist H., 1961) is written as

$$k = \frac{2(2 + x^2 - 2\sqrt{1 + x^2})}{x^2 \sqrt{1 + x^2}} \qquad (x = \frac{\delta\omega}{\gamma})$$

where

$\delta\omega$: FM amplitude
γ : halfwidth of the absorption line.

The theoretically maximized $k = 0.34$ is given by $x = 2.2$ (Arndt 1965). To obtain the highest sensitivity, we tuned the FM amplitude for the halfwidth of the air-broadened $2\nu_3$-band R(3) line of methane at 1 atm. The sensor output is obtained by the ratio between the 2*f* and *f* signals:

$$C_R = \frac{m}{2k\alpha_0} \cdot \frac{P_{2f}}{P_f}.$$

3 Detection-Limit Estimation

In the case of small optical depth, the 2*f* signal is much smaller than the *f* signal while the 2*f* and *f* current on PD have the same magnitude of noise. Thus we take only 2*f* component into account for detection-limit estimation. The 2*f* signal current (RMS) I_S is given by

$$I_S = \frac{1}{\sqrt{2}} s P_{DC} k \alpha_0 \cdot 2 C_R$$

where

s : sensitivity of the PD

The noise current I_N is given by

$$I_N = \sqrt{\left[s^2 (RIN) P_{DC}^2 + 2e(s P_{DC} + I_{dark}) + \frac{4 k_B T}{R_{sh}} \right] \frac{1}{2\Delta t}}$$

where

$-e$: charge of an electron (-1.602×10^{-19} C)
k_B : Boltzmann's constant (1.381×10^{-23} J/K)
RIN : related intensity noise of the laser
I_{dark} : dark current on the PD
T : absolute temperature of the PD
R : parallel resistance of the PD
Δt : phase - sensitive detection time constant.

If we define the detection limit C_R^{limit} as the range-

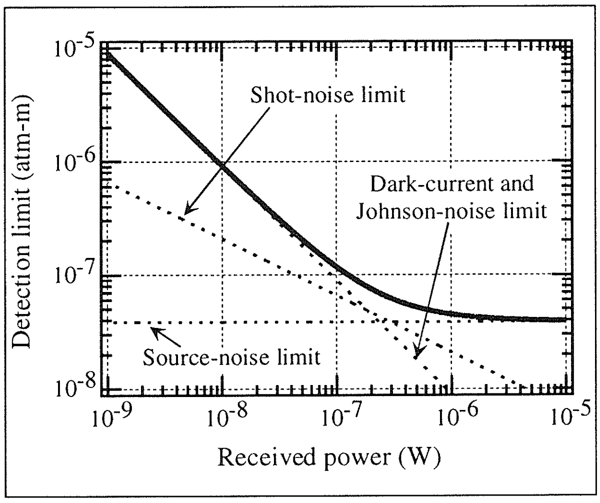


Figure 2. The theoretical detection limit vs. received power. The parameters used in calculation are as follows: (absorption coefficient) $\alpha_0 = 3.8 \times 10^1$ /atm/m, (laser) RIN = 10^{-12} /Hz, (FM) $k = 0.34$, (PD) sensitivity = 1.1 A/W; dark current = 100 nA; temperature = 300 K; parallel resistance = 500 kΩ, (phase-sensitive detection) time constant = 1 s.

integrated methane concentration of which the signal-to-noise ratio (SNR) equals unity, C_R^{limit} is given by

$$C_R^{\mathrm{limit}} = \frac{1}{k\alpha_0}\sqrt{\left[\frac{RIN}{2} + \frac{e}{sP_{DC}} + \frac{eI_{dark} + \frac{2k_BT}{R_{sh}}}{(sP_{DC})^2}\right]\frac{1}{2\Delta t}} \ .$$

Figure 2 shows the received-power dependence of the theoretical detection limit. The absorption coefficient at the center of the air-broadened $2\nu_3$-band R(3) line of methane is estimated $\alpha_0 = 3.8\times10^1$ /atm/m by HITRAN database (Rothman L. S. et al. 1992). In the case the received power is weak (< 100 nW), the detection limit is in inverse proportion to the received power.

4 Components of the Sensor

Figure 3 shows the compact remote methane sensor we have developed. The sensor consists of an optics unit (a) and a controller unit (b).

(a) The optics unit

The light source is a 1.65-μm InGaAsP DFB laser. The laser frequency is sinusoidally modulated at f = 10 kHz while the modulation-center is locked at the center of the $2\nu_3$-band R(3) line of methane. The injection current to the laser is set to 100-mA bias and 18-mA modulation (0-p). The laser light is coupled into an optical fiber (1.55-μm single mode), guided to a collimator of a grated index (GRIN) lens, and launched to a topographical target. The output power of the laser is about 5 mW at 100 mA. The diffuse reflection from the target is collected on the PD by a Fresnel lens (ϕ120mm, f255mm). The photodetector is an InGaAs PIN PD (ϕ1mm) and is packaged with a pre-amplifier.

(b) The controller unit

The controller unit not only controls the laser but also processes the signals. It has a digital signal processor (DSP) that performs the phase-sensitive detection with reference to f and $2f$. The time constant of the phase-sensitive detection is set to 1 s.

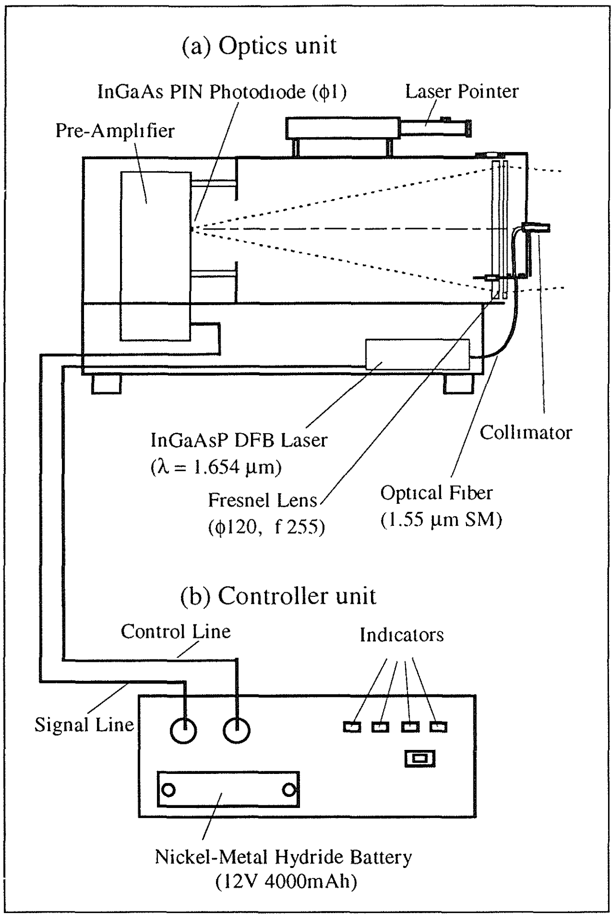
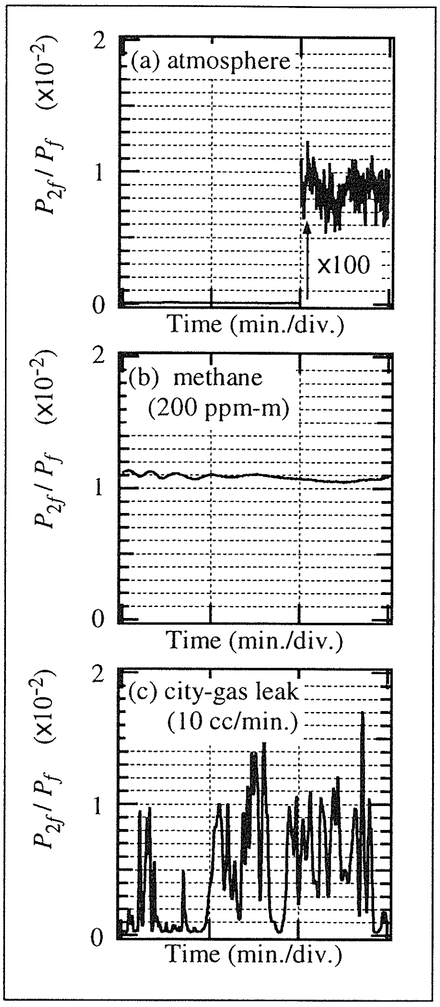


Figure 3. The schematic diagram of the compact remote methane sensor: optics unit (a) (ϕ130mm × L285mm, Wt.2.8kg) and controller unit (b) (W.200mm × D.250mm × H.90, Wt.3.0kg). The average consumption power in operation is 8W.

5. Experimental Results

First, we carried out the experiment with standard methane in an absorption cell in order to estimate the detection limit of the sensor. The target was barium sulfate ($BaSO_4$) that is widely known as a white standard with Lambertian surface, and was located at the range of 5 m with normal incident angle. In this condition, the received power P_{DC} was about 200 nW. Figures 4 (a) and (b) show the sensor output of atmosphere (a) and 200-ppm·m methane (1014-ppm methane in a 20-cm-long glass cell) (b). It should be noted that the output fluctuation in Figure 4(b) is due to an interference fringe caused by the cell window (*e.g.* Jin et al., 1997). The noise level was 1.4×10^{-5} and the signal level of 200 ppm·m methane was 1.1×10^{-2}. We estimate, therefore, the experimental detection limit is C_R^{limit} = 250 ppb·m. This is only about three times larger than the theoretical value in Figure 2. The optical depth corresponding to the experimental detection limit is $\alpha_0 \cdot 2C_R = 1.9\times10^{-5}$.

Next, we carried out the experiment of gas leak detection for city-gas containing about 80-vol.% methane to evaluate the performance of the sensor. The same target of $BaSO_4$ was used in the experiment. Figure 4 (c) shows the sensor output of 10-cc/min city-gas leak. This is the minimum flow rate to be detected in gas leak detection. Because gas was swung by slight wind, the output varied violently. However the sensor



Figures 4. Sensor outputs of atmosphere (a), 200-ppm·m methane (b), and 10-cc/min city-gas leak (c) (range: 5 m, target: BaSO$_4$, incident angle: normal, time constant: 1s). The received power was about 200

Table 1. The reflectivity of representative targets at incident angle of 60° ($\lambda = 1.6537$ μm) and corresponding detection limit of the sensor (range = 5 m).

Target	Reflectivity (/steradian)	Detection Limit (ppm·m)
barium sulfate	0.16	0.5
concrete	0.10	0.8
wood panel	0.07	1.1
marble	0.06	1.3
asphalt	0.03	2.7

detected such small leak with good SNR.

The high sensitivity obtained in the experiment promises the practical use with a realistic-material target and off-normal incident angle. Table 1 shows the reflectivity at incident angle of 60° of representative targets and corresponding detection limit. The detection limit is estimated by assuming it in inverse proportion to the received power.

6. Conclusion

We have developed a compact remote methane sensor using a TDL based on FM spectroscopy. The detection limit of the sensor with a 5 m barium-sulfate target (normal incident angle) was 250 ppb·m at the time constant of 1 s. The sensor detected city-gas leak at flow rate 10 cc/min.

References

Arndt R. (1965). Analytical Line Shapes for Lorentzian Signals Broadened by Modulation. *J. Appl. Phys.* **36** 2522-24

Bobin B. (1972). Interprétation de la bande harmonique $2\nu_3$ du Méthane $^{12}\mathrm{CH}_4$ (du 6890 à 6107 cm^{-1}). *J. Phys.* (Paris) **33** 345-52

Jin W., Xu Y. Z., Demokan M. S., and Stewart G. (1997). Investigation of interferometric noise in figer-optic gas sensors with use of wavelength modulation spectroscopy. *Appl. Opt.* **36** 7239-46

Rothman L. S. et al. (1992) The HITRAN molecular database: edition of 1991 and 1992. *J. Quant. Spectrosc. Radiat. Tranfer. 48* 469-507

Uehara K. and Tai H. (1992) Remote detection of methane with a 1.66 μm diode laser. *Appl. Opt.* **31** 809-14

Wahlquist H. (1961) Modulation Broadening of Unsaturated Lorentzian Lines. *J. Chem. Phys.* **35**