# Remote Measurements of Atmospheric Trace Molecules with Lasers(レーザーによる大気中微量分子の遠隔測定に関する研究)

| | |
|---|---|
| 氏　　　　　名 | ムスタファ・ハムザ・イブラヒム・サイド・エル・アヘル<br>MOSTAFA HAMZA IBRAHIM SAYED EL-AHL |
| 授　与　学　位 | 工　学　博　士 |
| 学位授与年月日 | 昭和 56 年 5 月 13 日 |
| 学位授与の根拠法規 | 学位規則第 5 条第 1 項 |
| 研究科，専攻の名称 | 東北大学大学院工学研究科<br>（博士課程）電子工学専攻 |
| 学位論文題目 | Remote Measurements of Atmospheric Trace Molecules with Lasers<br>（レーザーによる大気中微量分子の遠隔測定に関する研究） |
| 指　導　教　官 | 東北大学教授　稲場　文男 |
| 論文審査委員 | 東北大学教授　稲場　文男　　東北大学教授　高橋　正<br>東北大学教授　柴田　幸男　　東北大学教授　堀江　忠児 |

# 論　文　内　容　要　旨

## CHAPTER 1 INTRODUCTION

The laser rader has been achieving great attention as a new tool for remote monitoring the various properties of the environmental atmosphere. So far a variety of capabilities of the laser rader technique has been demonstrated. In order to meet the severe requirements as practical and valuable remote monitoring instrumentations, however, the laser radar needs further improvement in the sensitivities, applicabilities and other engineering problems.

This thesis was intended to study and extend fundamental and application techniques of the laser remote measurements of atmospheric trace molecules based on the differential absorption method. The main efforts were concentrated in the design, development and construction of two-wavelength oscillation $CO_2$ lasers which are of fundamental importance for the application in the differential absorption laser radar system. The long-path differential absorption monitoring systems were developed using the two-wavelegth oscillation $CO_2$ lasers for high sensitive measurement of air pollution molecules.

Furthermore, the differential absorption laser radar system employing the topographic targets was also developed using the infrared heterodyne detection technique for the high sensitive remote measurement of the atmospheric trace molecules.

The design and experimental results of these systems were presented and discussed in this thesis which was divided into seven chapters.

## CHAPTER 2 LASER RADAR TECHNIQUES FOR REMOTE MEASUREMENTS OF ATMOSPHERIC TRACE MOLECULES

The laser radar technique has made it possible to realize the single-ended measurement of optical signals backscattered from materials existing in the atmosphere. By applying spectroscopic analytical techniques in the laser radar, qualitative and quantitative informations about atomic and molecular constituents and their concentrations are obtained. In this chapter, the basic characteristics of the laser radar technique was reviewed and discussed. Three different laser radar systems based on Raman scattering, resonance fluorescence and absorption methods have been discussed and compared on the capabilities and limitations of their performances. It has been clear that the differential absorption method offers the best sensitivity incorporating with a long-path measurement of trace atmospheric molecules and requires least laser output powers.

Characteristics and performances of the laser sources for the laser radar applications were summarized. For the differential absorption laser radars in the infrared region where most molecules have strong vibration-rotation absorption spectra, the $CO_2$ laser can be used effectively because of its high power, high efficiency and step-tunable features in 9.6 μm and 10.6 μm bands.

## CHAPTER 3 MEASUREMENTS OF ABSORPTION COEFFICIENT AND DIFFERENTIAL ABSORPTION OF MOLECULES WITH $CO_2$ LASERS

For the estimation of the sensitivity and potentiality of the differential absorption remote sensing method with lasers, detailed information on the strength and the nature of the absorption spectra of the molecule to be monitored is basically important. In this chapter, the absorption coefficients of some atmospheric trace molecules were measured and tabulated for the use in the later parts of this thesis.

We have analyzed that in the lower atmosphere the spectral lines are pressure broadened to about 0.1 $cm^{-1}$ for the spectral width and there is a great probability of overlapping with oscillation lines fromsdiscretely tunable $CO_2$ lasers.

Absorption coefficients of $C_2H_4$ and $NH_3$ molecules were measured in the pressure broadened region of one atmospheric air pressure using the $CO_2$ laser spectrometer at many oscillation branch wavelengths.

Differential absorption coefficients of $C_2H_4$ molecules were measured by developing the dual $CO_2$ laser differential absorption spectrometer and the best selection of the $CO_2$ laser line pair for the long-path differential absorption measurement of $C_2H_4$ molecules in the atmosphere has been found.

## CHAPTER 4 TWO-WAVELENGTH OSCILLATION $CO_2$ LASER USING GRATING-ANGLE MODULATION FOR DIFFERENTIAL ABSORPTION MEASUREMENTS

In this chapter, the basic relations for a new technique of the long-path differential absorption scheme were derived. The two-wavelength and output power-balanced oscillation in a single $CO_2$ laser oscillator has been realized using both the control techniques of the grating-angle modulation and the cavity length change by means of PZT driven mirror.

This new scheme of $CO_2$ laser oscillation is particularly suited for the differential absorption measurement of low concentration atmospheric trace molecules. Incorporating this performance of the $CO_2$ laser, the phase sensitive detection method was applied to derive accurately the signal difference between the received powers at the two wavelengths transmitted into the atmosphere.

This method offers a significant simplification of the detection system against the conventional differential absorption method and also an effective improvement in the detection sensitivity by reducing not only the mechanical and optical instabilities but also the atmospheric scintillation noise.

As a demonstration of the usefulness of the technique described in this chapter, $C_2H_4$ molecules have been detected experimentally both in a calibration cell and also in the exhaust of a combustion engine. Minimum detectable absorption was approximately 0.8 % in this experiment.

The experimental set-up of the differential absorption spectrometer using the two-wavelength oscillation $CO_2$ laser is shown schematically in Fig. 1. Fig. 2. shows the oscilloscope trace of the $CO_2$ laser output and the driving voltage with a square waveform applied to the electromagnet. The alternate oscillation of the two wavelengths at P(14) and P(16) branches in 10.6 $\mu$m band of the $CO_2$ laser used for the detection of $C_2H_4$ molecules is shown in this Figure with almost balanced output powers. As an example, Fig. 3. shows the result of the measurement of concentration of $C_2H_4$ in the exhaust of a small combustion engine during different operating conditions.

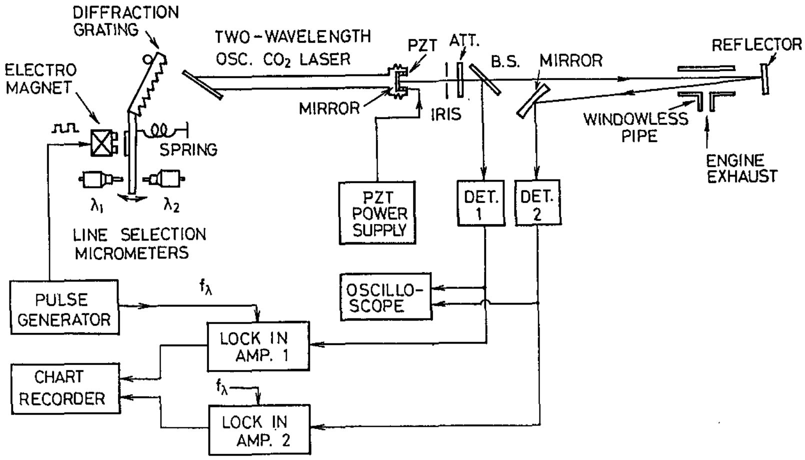


Fig. 1 Block diagram of the two-wavelength oscillation $CO_2$ laser spectrometer used for the long-path differential absorption measurement of molecular species.

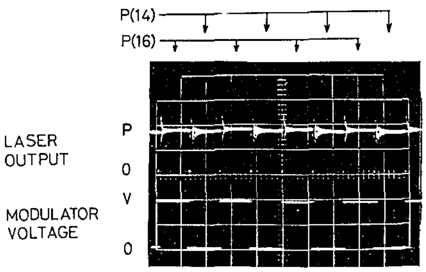


Fig. 2 Oscilloscope trace of the $CO_2$ laser output (upper trace) and the modulator output voltage (lower trace).

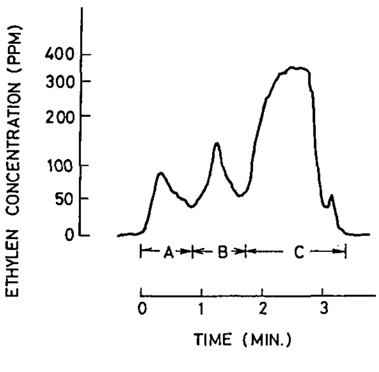


Fig. 3 Detection of $C_2H_4$ molecule in the exhaust of the motor cycle engine. Region A is ignition and idling, B is medium speed (3500 RPM) and C is high speed (7000 RPM) running conditions. Lock-in amplifier time constant is 10 sec.

# CHAPTER 5 TWO—WAVELENGTH OSCILLATION $CO_2$ LASER USING MIRROR—ANGLE MODULATION FOR HIGH SENSITIVE DIFFERENTIAL ABSORPTION MEASUREMENTS

Although the grating-angle tilting method described in the previous chapter has offered higher sensitivity than the previous methods, further improvement in the sensitivity of the differential absorption scheme is needed for the accurate measurement of trace molecules.

In this chapter, another two-wavelength oscillation $CO_2$ laser has been developed using the mirror-angle modulation method for further improvement of the detection sensitivity in differential absorption measurements. The design and performance of this $CO_2$ laser was presented and the results of applying this laser to the long-path differential absorption system were discussed and summarized.

The stable high-frequency modulation and the automatic output power-balancing for the two-wavelength oscillation have been realized. From these features we could improve the detection sensitivity of the long-path differential absorption measurement. Also they allow us to provide a long term and stable operation of the differential absorption system employing this $CO_2$ laser.

Using this long-path differential absorption system, $C_2H_4$ molecules have been detected experimentally both in a calibration cell and also in the open atmosphere. Minimum detectable absorption was found to be nearly 0.1% in the cell experiment and 0.3% in the long-path outdoor experiment.

Fig. 4 shows the block diagram of the experimental system using the power—balanced, two—wavelength oscillation $CO_2$ laser described in this chapter, for

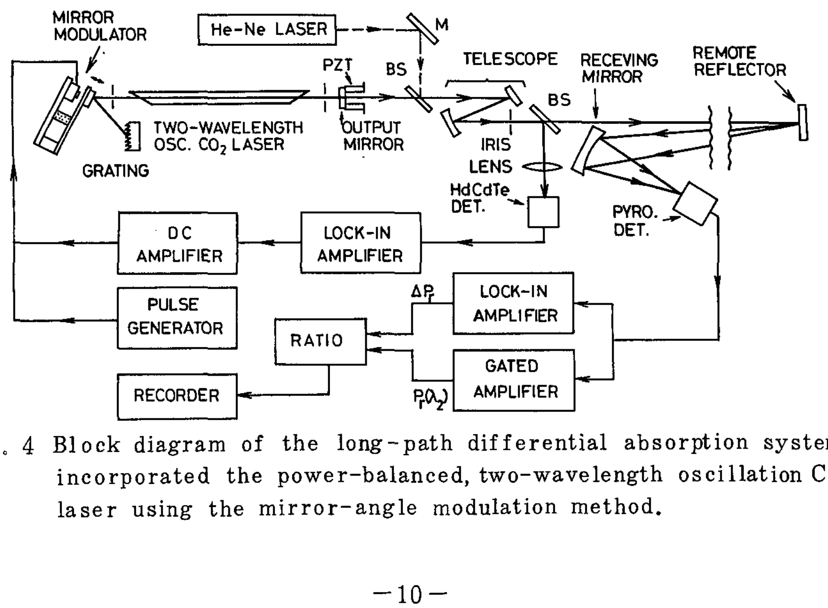


Fig. 4 Block diagram of the long-path differential absorption system incorporated the power-balanced, two-wavelength oscillation $CO_2$ laser using the mirror-angle modulation method.

the long-path differential absorption measurement of $C_2H_4$ molecules. Fig. 5 is an example of oscilloscope trace illustrating the power-balanced, two-wavelength oscillation in the P(14) and P (16) branches of the $CO_2$ laser at the modulation frequency of 100 Hz. Fig. 6 shows an example of the experimental result of the differential absorption measurement of $C_2H_4$ molecules diffused in the atmosphere from the exhaust of a running motor cycle engine.

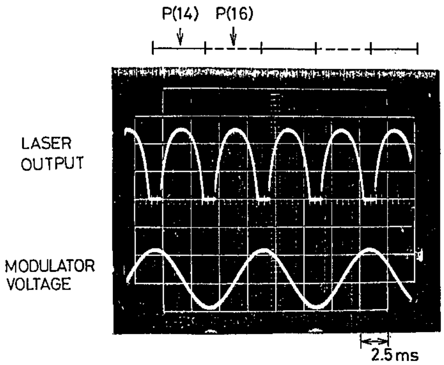


Fig. 5 Oscilloscope trace of the power - balanced, two - wavelength $CO_2$ laser oscillation in the P(14) and P(16) branches realized by the automatic power-balance controller at the modulation frequency of 100 Hz.

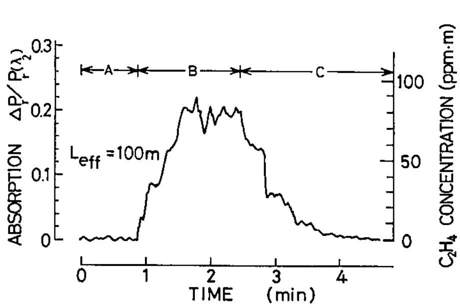


Fig. 6 Differential absorption detection of $C_2H_4$ molecules diffused in a long - path open atmosphere. In region B, a motor cycle engine was started and run near the optical path, while region A indicates condition before the drive and in region C, the engine was stopped. Detection time constant is 3 sec.

## CHAPTER 6 HETERODYNE $CO_2$ LASER RADAR SYSTEM FOR REMOTE DIFFERENTIAL ABSORPTION MEASUREMENTS OF ATMOSPHERIC TRACE MOLECULES

More flexible and wide applications of the differential absorption measurement are expected by utilyzing the topographic targets as the reflector. In this case the molecular concentration in various directions, distances and places could be measured and the two dimentional mapping of the concentration becomes possible.

In this chapter, the infrared heterodyne detection laser radar system has been developed incorporating the FM—CW technique for the differential absorption measurement of atmospheric trace molecules using topographic targets as the reflectors. The heterodyne detection method was found to give higher detection sensitivity even using a low-power $CO_2$ laser compared with the direct detection method conventionally used in the infrared.

The FM—CW heterodyne laser rader has been constructed using a compact waveguide $CO_2$ laser as the transmitter for which the frequency modulation by the laser cavity length modulation technique was applied. The range of the target was measured from the beat signal frequency between the tranmitted and recieved laser beams.

Furthermore, a simple and compact differential absorption laser radar system incorporating the FM—CW heterodyne scheme has been realized based on the development of the power-balanced, two-wavelength oscillation method by means of the grating-angle modulation technique. In the long-path experiment to observe $C_2H_4$ molecular concentrations in the atmosphere using a building wall as the reflector, the minimum detectable absorption of approximately 0.5% was achieved.

Fig. 7 shows the block diagram of the $CO_2$ laser radar system. Fig. 8 shows a typical example of the beat signal spectrum observed for the backscatter from a building wall as a topographic target. Fig. 9 shows an oscilloscope trace of the power-balanced, two-wavelength oscillation output of the $CO_2$ waveguide laser and the driving voltage applied to the grating-angle modulator. The P(14) and P(16) branch pair in the 10.6 μm was selected for the alternate oscillation for the detection of $C_2H_4$ molecules. Fig. 10 shows a typical result of the long-path measurement of $C_2H_4$ molecules in the atmosphere.

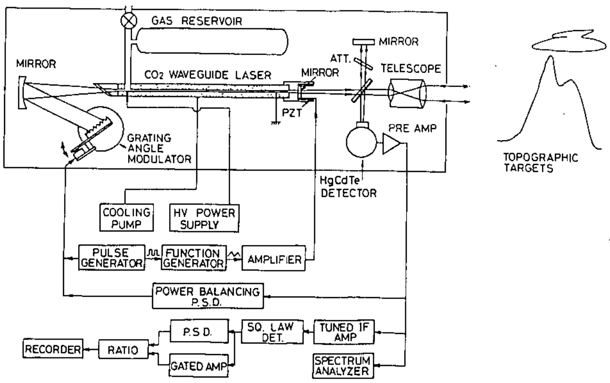


Fig. 7 Block diagram of the FM—CW heterodyne laser radar system with a power-balanced, two-wavelength oscillation $CO_2$ laser for differential absorption measurements using topographic targets.

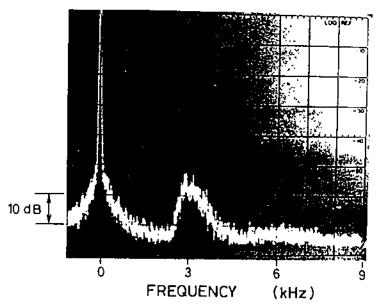


Fig. 8 Beat signal spectrum centered around 3 kHz observed for a target at 200 meter range. Spectral width of 300 Hz corresponds to 20 meter range resolution.

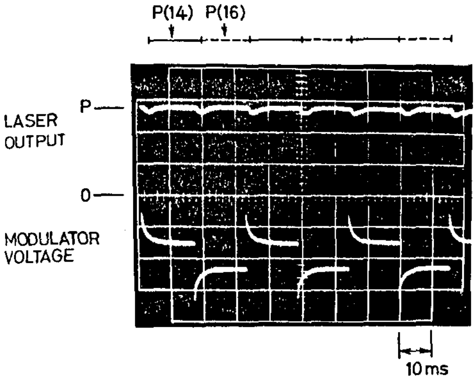


Fig. 9 Oscilloscope trace of the alternate two-wavelength oscillation at P (14) and P (16) branches in the $CO_2$ laser using the grating-angle modulation method.

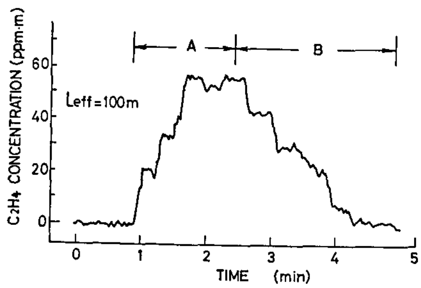


Fig.10 Remote measurement of $C_2H_4$ molecules by long-path differential absorption method using the FM—CW heterodyne $CO_2$ laser radar. In region A, a two-stroke motor cycle engine was driven along the optical path and in region B, the engine was stopped. Detection time constant is 3 sec.

The laser radar system described in this chapter is compact and easily movable by loading on a small car that measurements in various directions and places can be expected. Also, many other molecules such as $O_3$, $NH_3$, Freon, $CO_2$ and $H_2O$ could be measured with this FM—CW heterodyne $CO_2$ laser radar system by properly selecting the oscillation wavelengths.

## CHAPTER 7 SUMMARY AND CONCLUSIONS

This thesis has described the design and performance of alternate two-wavelength oscillation $CO_2$ lasers based on the wavelength selective modulation techniques, and their applications to the high-sensitive remote differential absorption measurements of atmospheric trace molecules.

The results obtained in this thesis may give significant contributions to the field of laser engineering and the laser radar technology mainly in remote sensing applications. Further research and development of tunable lasers and laser radar systems will be able to find many new applications in active remote sensing technology relevant to both research and practical purposes.

# 審 査 結 果 の 要 旨

レーザー光を大気中に発射して微量成分分子の遠隔計測を行うレーザー･レーダーは，レーザーのすぐれた電子工学的応用の一つとして，近年急速に研究開発が進められている。この間，高出力レーザーをはじめ各種の光検出技術や信号処理法などが開発され，レーザー・レーダーの性能向上に貢献して来たが，これらの技術の総合的な検討にもとづいて，大気中に拡散した微量な車輌排気ガス濃度などのレーザーによる実時間遠隔測定法の開発が実用上強く望まれていた。

著者はこのような観点から，赤外域$CO_2$レーザーの発振線の一部がエチレンの吸収スペクトルと一致することに注目し，装置の設計，試作を行って，長光路にわたるレーザー光の吸収量の精密な計測によってエチレンの濃度の遠隔測定が可能であることを明らかにした。本論文はその成果をとりまとめたもので，全編7章よりなる。

第1章は総論である。第2章では，レーザー・レーダーの種々の分光的動作方式と装置構成の検討を行い，差分吸収を利用する方式が実用上十分な感度を持つことを論じている。

第3章では，エチレン分子の赤外域吸収バンド内のスペクトルが$CO_2$レーザーの10.6 $\mu$m 帯の発振線の一部と合致することを測定し，その基本的な分光特性を調べて，本測定法の物理的基礎を明確にしている。

第4章では，大気中の微量エチレン濃度の遠隔測定のためには，吸収波長とそれに近接する吸収の少ない波長の二波長発振$CO_2$レーザーの実現が必要であることに着目して，レーザー共振器の一部に回析格子を使用し，その角度を変調すると共に，二波長出力が交互に等しくなるような出力制御系を設けた新しい方式を考案して，室内実験によってその有用性を実証している。

第5章では，さらに前章で開発した同一出力の二波長発振$CO_2$レーザーの高性能化をはかるために，変調制御方式に独特の改良を加え，排気ガス中で実際に変動しているエチレン濃度の実時間遠隔計測を確認している。

第6章では，本レーザー・レーダー装置の小型，軽量化による移動遠隔測定を可能にするために，光導波路型の小型$CO_2$レーザーを効果的に利用した連続発振・周波数変調方式ヘテロダイン検波法をはじめて実現し，そのすぐれた動作特性を解明すると共に，長光路中に拡散している車輌排気などによる微量エチレン濃度の実時間計測に成功している。

第7章は結論である。

以上要するに，本論文は赤外域$CO_2$レーザーを利用して大気中エチレン濃度の遠隔計測を行う長光路差分吸収測定装置を研究，開発し，幾つかの興味ある知見を得たもので，電子工学ならびにレーザー工学の発展に寄与するところが少なくない。

よって，本論文は工学博士の学位論文として合格と認める。