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Fast Spectrometry System with Using Tunable Laser Diode

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Synopsis

Basic problems are discussed about a local, real-time and very sensitive air-pollution monitoring using a laser diode. The method employs the second derivative spectrometry replacing the incoherent light source and mechanical choppers in a traditional method with the laser diode and a newly developed electronic system. Etalon fringes at this system becomes the dominant noise source and its statistic and dynamical features are to be understood.

Numerical examination was done for the optimal width of optical frequency modulation. A very fast measurement of weak absorption spectra became possible by the development of lock-in-amplifiers with finite integration time instead of a low-pass filter. Spectra of methane in 7.6 μm region and a time-dependent spectrum of the etalon fringe are shown.

1. Introduction

Derivative spectroscopy has found many applications in both laboratories and field uses taking advantage of its high sensitivity and high specificity.

Williams and Hager[1] constructed a portable derivative spectrometer for a point air-pollution monitoring. Its theoretical problems are also reported by Hager and Anderson[2]. They succeeded in detection of some hundreds ppm of NH_3, No, and SO_2, using ultraviolet

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spectral region. Strojek *et al.*[3] could detect 230 ppb•m of So_2 by employing a 88 cm-long cell, which showed a possibility for the air-pollution measurement with real-time and point monitoring features. Detailed theoretical analysis for the errors induced by the inherent nonlinear characteristics in the derivative spectrometry was studied by O'Haver and Green[4]. They concluded that the derivative spectrometry gave the lowest total errors in quantitative measurement in most of the cases studied.

Applications of the derivative spectrometers with using a combination of a grating spectrometer and vibrating slits or mirrors are limited to several polluting gas species, for instance, NH_3,NO, NO_2, and SO_2. It is because the spectral resolution of a portable grating spectrometer is not sufficient for taking the rotational-vibrational structures of most polluting gases that only the enough broad electron-transition bands for short spectrometers are utilized.

Advent of lead-salt diode has invoked a very high resolution spectroscopy in the infrared region because of its very narrow spectral width and fine tunability. Hinkley *et al.*[5] could resolve individual transitions of the vibration-rotation band of SO_2 in the 8.7 μm region, and could determine pressure broadening coefficients directly from the high resolution data. Aronson *et al.*[6] showed that ν_1 vibration of SO_2 can be resolved, which had been impossible by conventional monochrometers.

Laser technology has already been used in remote air-pollution monitoring[7]. The introduction of laser diode has added more excellent portability and lighter weight features to this technology. Some attempts have been made in long-path monitoring of atmospheric pollutions [8] [9] using laser diodes. Atmospheric turbulence could be overcome by taking the advantage of the rapid frequency modulation of the laser emission.

Reid *et al.*[10] [11] succeeded in detecting 5 ppb of SO_2 and NO_2 at atmospheric air, and 0.1 ppb of NH_3 by using the lead-salt laser diodes along with 300 m path-length in a White cell. The second derivative method is there employed taking advantage of the quick frequency tunability resulting in an dramatic noise reduction mainly caused by mechanical vibrations.

These high sensitivities owe, in part, to very long time constant of the lock-in-amplifier (LIA) employed. This result may be an obstacle to a real-time measurement of air-pollution in the human-environment. The development of a quicker method is, therefore, expected.

The authors have studied a point real-time air-pollution monitoring method for years. Their first attempt was made in Raman scattering method, which failed by the cause of its inherent low sensitivity. Then a resonant absorption method for a pulsed dye laser was employed, which resulted in a conjectured attainable highest sensitivity data, but a real measurement was not made by the cause of its difficulties in realizing a portable apparatus and of a drawback of the pulsed operation in rather slow repetition rate.

These problems, however, were resolved by using compression electronic circuit technology and a numerical spectrum processing techniques, by which one can detect a spectrum of very weak attenuation out of noise, discriminate a specific spectrum of a gas, and calculate its gas density with a microcomputer [12][13].

Meanwhile, laser technology in lead-salt diode has been founded also in Japan, and enough reliable diodes can be supplied by Fujitsu Laboratories Ltd.[14]. Being supplied with their test products, the authors have studied fundamental problems for the sensitive, real-time, and point monitoring for air-pollution.

For a sensitive point monitoring, it has already been pointed out that inevitable and undesirable etalon fringes are the dominant noise source [11]. The improvement of this method for a practical use depends on a development of a technique to suppress the fringes. This paper is a report on our development of the basic technologies for the monitoring method. Efforts were made at the development of the electronic system which can give a second derivative spectrum normalized by the power in a short period in order to understand the dynamics of etalon fringes.

2. Preliminary Analysis

2.1 Second Derivative Spectrum

The principal process of an absorption spectrometry using a laser diode is expressed by an equation,

$$P(\nu) = P_0(\nu)e^{-\tau(\nu)}, \tag{1}$$

where the notations are

ν : frequency of the light,
τ : attenuation for the light,
$P(\nu)$: optical output power,
P_0 : optical incident power,

and then, ν, is modulated by a sinusoidal waveform as

$$\nu = \nu_0 + \Delta\nu\cos\omega t, \tag{2}$$

where ν_0 is the center frequency and $\Delta\nu$, the amplitude difference of frequency.

Assuming $\Delta\nu$ is small enough, the optical power, $P(\nu)$, can be expanded up to second harmonics involving the second order with respect to $\Delta\nu$ in the following Eq.(3).

$$P(\nu) \simeq P(\nu_0) + \frac{1}{4}P''(\nu_0)\Delta\nu^2 + P'(\nu_0)\Delta\nu\cos\omega t + \frac{1}{4}P''(\Delta\nu)^2\cos 2\omega t \ , \tag{3}$$

where the notations are

$$P'(\nu_0) : \frac{\partial P(\nu_0)}{\partial \nu}$$

$$P''(\nu_0) : \frac{\partial^2 P(\nu_0)}{\partial \nu^2} \ .$$

The amplitude of the second harmonic term can be obtained by employing a LIA and the $P''(\nu)$ term is described from Eq.(1) as

$$P''(\nu) = [P_0''(\nu) - 2P_0'(\nu)\cdot\tau' + P_0(\nu)\cdot\{(\tau')^2 - \tau''\}]e^{-\tau}, \tag{4}$$

where the notations are

$$\tau' : \frac{\partial \tau}{\partial \nu}$$

$$\tau'' : \frac{\partial^2 \tau}{\partial \nu^2} \ .$$

2.2 Distortion

In the following, magnitude of τ is assumed to be very small as

$$\tau << 1 \ . \tag{5}$$

This condition is effective for a measurement of very small concentrations of polluted gas species in the air.

In order to eliminate an effect of long term change on the diode output, a normalization process is necessary, and its result, a 2f-spectrum, S_{2f}, is described as

$$S_{2f} = \frac{1}{4}\{\frac{P_0''(\nu)}{P_0(\nu)} - \frac{P_0'(\nu)}{P_0(\nu)}\tau' + (\tau')^2 - \tau''\}e^{-\tau}\cdot\Delta\nu^2 \ . \tag{6}$$

The term that linearly corresponds solely to the attenuation is only τ'' term and others are suffered from the $P_0(\nu)$ or $P_0'(\nu)$ values which may be affected by low frequency ($1/f$) noise or drift in the laser output.

It is, therefore, necessary to examine how much appendix terms affect on the S_{2f} in comparision with the τ'' term. For this purpose, some assumptions are put. The $P_0(\nu)$ changes linearly to $\delta\nu$, a deviation of ν from the center frequency ν_0, as

$$P_0(\nu) = P_0(\nu_0) + P_0'(\nu_0)\delta\nu \tag{7}$$

and the spectrum of $\tau(\nu)$ is an isolated line of Lorenzian form with its center at ν_0 and the half width at half maximum (HWHM) of $\tau(\nu_0)$, Γ, viz.,

$$\tau(\nu) = \frac{\Gamma^2}{(\nu-\nu_0)^2 + \Gamma^2}\,\tau_0 \ , \tag{8}$$

where τ_0 is the strength of τ at the center. Shapes of the τ, τ', and τ'' are shown in Fig.1. An interference term $P_0'\tau_0'/P_0$ may be the largest at $\delta\nu = \Gamma/3$ and its magnitude should be compared with the τ'' value at $\delta\nu=0$.

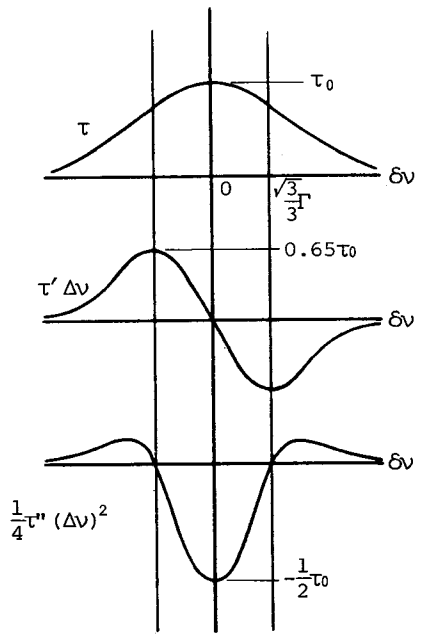

Fig.1 Fundamental, 1st-derivative and 2nd-derivative spectra of a Lorenzian line shape. The $\Delta\nu$ is taken to be the HWHM, Γ.

2.3 Etalon Fringe

It has been reported that the etalon fringe is the main factor which limits the signal to noise ratio(SNR) in the taken spectrum. The etalon fringe takes place due to a couple of opposite reflecting surfaces across the optical path. This has not been cleared when a white and incoherent light sourse was employed. Coherence of the laser causes this phenomenon. The magnitude of the etalon fringe in terms of the attenuation is of the order of the secondary reflecting light, $R^2P_0(\nu)$, which is convinced referring Fig.2, where R is the reflecting ratio of the surface.

The amplitude of the etalon fringe in the LIA output, however, depends on the amplitude of the frequency modulation, $\Delta\nu$. Denoting the etalon-fringe spectrum as

$$\varepsilon(\nu) = \bar{\varepsilon}\cos(2\pi\nu/p), \tag{9}$$

where $\bar{\varepsilon}$ and p stand for the amplitude and a pitch of the fringe, respectively. The 2f-spectrum of the LIA output, S_{2fet}, is obtained analytically as

$$S_{2fet} = 2\pi\bar{\varepsilon}\cdot J_2(2\pi\Delta\nu/p) \ . \tag{10}$$

where $J_2(2\pi\Delta\nu/p)$ is the Bessel's function.
This value is further expressed in an approximation as

$$S_{2fet} \sim 2\bar{\varepsilon}\sqrt{\frac{p}{\Delta\nu}}\cos\left(2\pi\frac{\Delta\nu}{p}+\frac{3}{4}\pi\right) \ , \tag{11}$$

for large values of $\Delta\nu/p$.

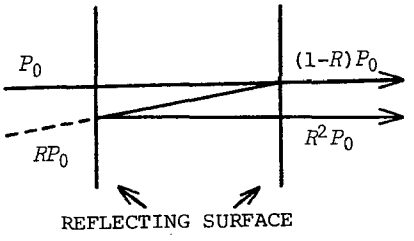

Fig.2 Etalon fringe takes place due to the two reflecting surfaces. The power of the fringe is of the order of the secondary reflecting light, R^2P_0.

2.4 Optimal Modulation Amplitude

The 2f spectrum of the LIA output becomes different from the value given by Eq.(6) when $\Delta\nu$ is large. Fig.3 gives a result of a numerical analysis about the magnitude of the 2f spectrum at the frequency center expressed as a function of $\Delta\nu$. Assuming the same absorption spectrum with Eq.(8), the maximum 2f amplitude is attained at $\Delta\nu/\Gamma=2.2$. The amplitude of the frequency modulation should be chosen at this value. The optical system should be designed so that the etalon spacing, that cannot be got rid of, should be designed so that the condition of either

$$\Delta\tau/p >> \Gamma \text{ or } \Delta\tau/p << \Gamma \tag{12}$$

should be hold.

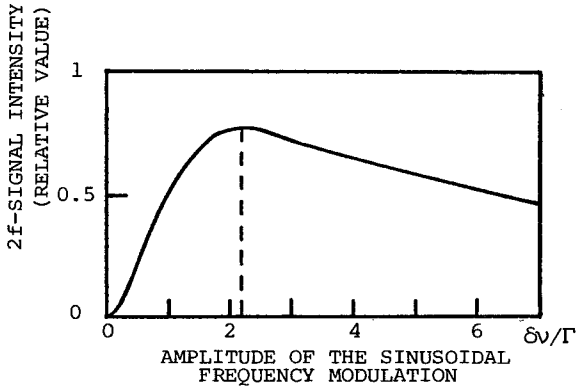

Fig.3 Magnitude of the 2f signal for various amplitude of the frequency modulation. The maximum output is attained at $\delta\nu/\Gamma=2.2$.

3. Experiments

3.1 Experimental Setup

The block diagram of the electric signal system is given in Fig.4. A driving current, I_D, of the laser diode consists of three terms as

$$I_D = \{I_0 + \Delta i\cos(2\pi ft)\}u(t) \ , \tag{13}$$

where I_0 is a staircase waveform whose temporal value corresponds to the optical frequency of the spectra. The duration of a step has a nomenclature " *time slot* ", and the whole system including the LIA operates according to a prescribed sequence in the time slot. The second term, $\Delta i\cos 2\pi ft$, stands for a sinusoidal amplitude modulation with small amplitude. The multiplying factor, $u(t)$, is employed to express the blanking period when the drive current is enforced to null with analog switch circuit of V-MOS transistors. The waveform of I_D is given in Fig.5 along with parameters, which was determined from experiments for the dynamical response of the laser diode[15]. The time sequence of I_D is controlled by a clock signal which is synchronized to the 60 Hz power line voltage. By this, the " *ham* " problem of the high-gained signal processing are circumvented.

Output of the infrared detector (ir-detector) is connected to two LIAs. One is for 2f signal, and the other for power monitoring. Different from the LIA commonly used, the output of the phase-sensitive detector is integrated for a finite period until it is cleared, instead of being fed to a low-pass filter.

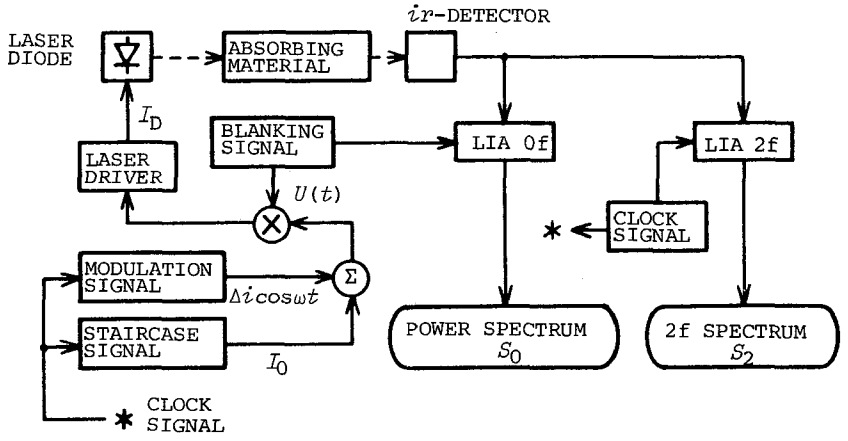

Fig.4 Block diagram of the total electronic circuits. The LIA cooperates with the blanking signal generator to obtain the laser output. Both LIAs are featured by its finite integration period instead of a low-pass filter in a usual configuration.

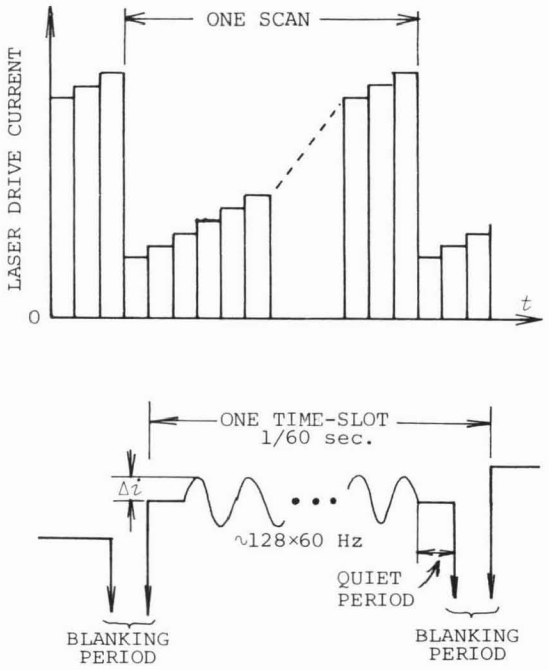

Fig.5 Waveform of the laser drive current. The current is of a staircase profile with sinusoidal modulation of a small amplitude. The blanking period is for the power measurement. The quiet period is necessary to ensure a correct power measurement.

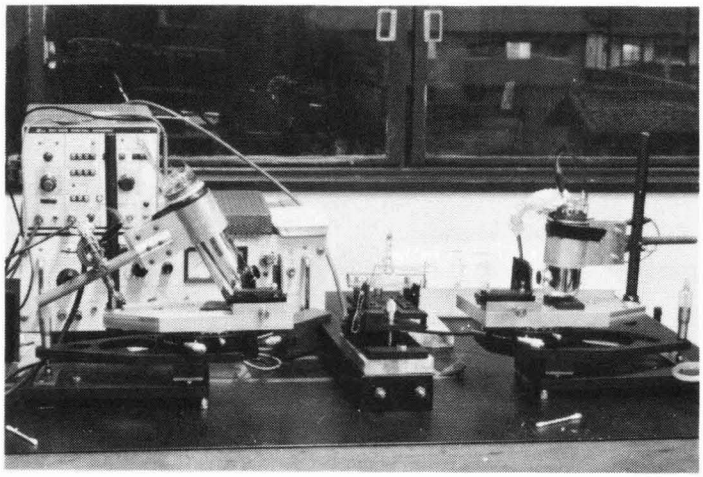

Fig.6 Optical setup of the system.

The period of integration is synchronized to the time slot. This configuration allows a very rapid scanning of the driving current without a fear of spectral distortion. The LIA for power monitoring can detect the discontinuous voltage change of the ir-detector output at the beginning instant of the laser blanking. With this power monitoring circuit a troublesome mechanical chopper was replaced, which also helped the rapid scanning. Fig.6 shows the optical configuration.

3.2 Results of Experiments

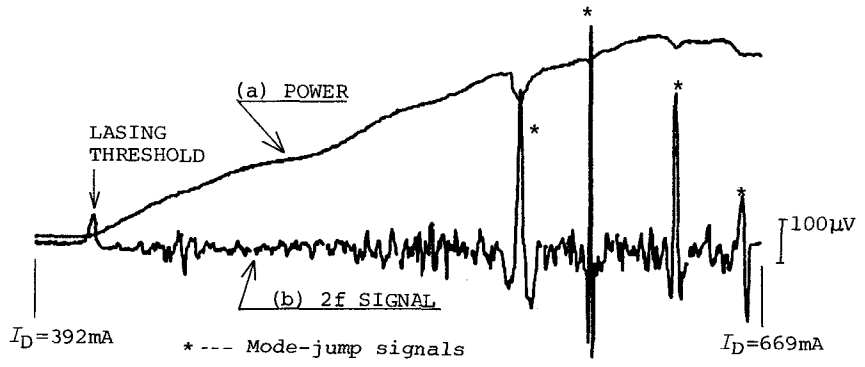

Fig.7 Spectra of laser power and its 2f spectrum. Intense etalon fringes as well as the mode-jump signals are clearly observed.

Fig.7 is a result with wide scanning width of I_d involving no specific absorbing material. The trace (a) is the power spectrum. Lasing occurs only above the threshold and many output discontinuities are observed. Each discontinuities corresponds to borders of adjacent lasing modes to which discontinuous optical frequency band is associated. The discontinuity is exaggerated by the trace (b), the 2f signal spectrum. The tuning rate defined by $d\nu/dI_D$ can be found on Fig.8, in which the power spectrum of the laser light passed through a 5 mm thick germanium etalon plate is shown. One cycle of its undulation corresponds to 0.25 cm^{-1}.

Fig.9 is a spectrum of pure methane with 3 Torr concentration. Many distinguished line spectra are observed. The line width increases with a higher pressure, and a cluster of lines is resolved into a band as is shown in Fig.10, which is of 958 ppm at atmospheric air pressure.

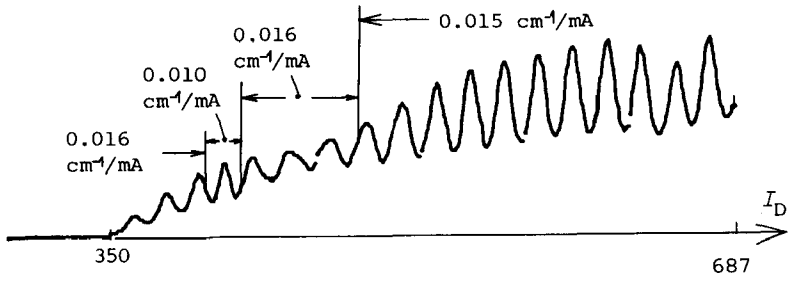

Fig.8 Spectrum of the Ge etalon plate with 5 mm thickness. The tuning rate of the diode laser are found. The rates are not the same between different modes.

Fig.9 Trace of 2f spectrum of methane, 3 Torr, 100%.

The undesirabl etalon fringe is observed as Fig.11 when a gain of the LIA is increased up to its maximum. The traces were taken every 1.67 second, and it is found that the fringes wander slowly. An isolated line of 0.1 Torr pure methane is taken simultaneously as an absolute frequency marker.

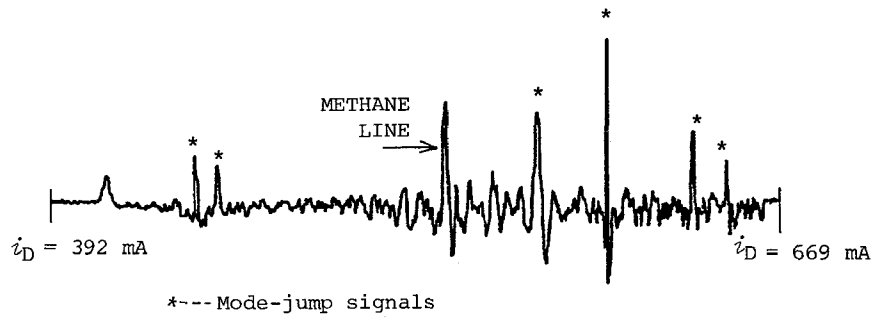

Fig.10 Trace of 2f spectrum of methane of 958 ppm at atmospheric air-pressure. A cluster of lines is collected into a rather broad line.

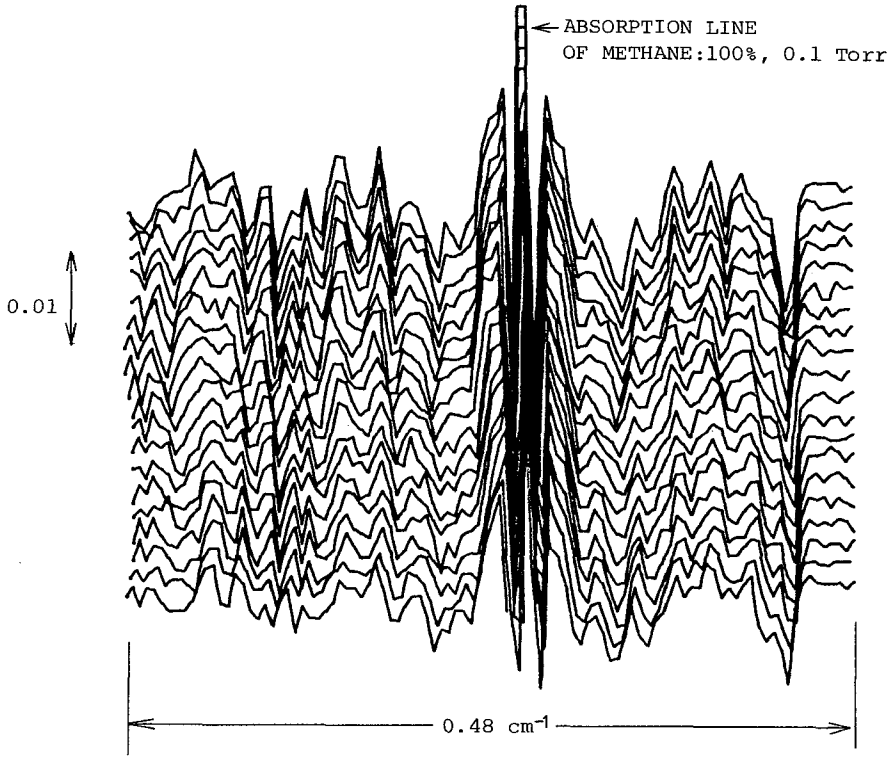

Fig.11 Sweep scans of etalon fringe spectrum. Every trace is over 1.67 second and successively follows the preceding scan. In order to fix the optical frequency axis, an isolated and very sharp line of methane with 0.1 Torr is overlapped.

4. Concluding Remarks

A new system of ir-spectrometry using a diode laser and a ir-detector has been developed. The fully electronized configuration of the driving method for the laser along with the new LIA configuration with finite integration period made it possible to obtain 2f and power spectrum in a far shorter period than it was by the traditional spectrometer-chopper composition.

A dynamical feature of etalon fringe has become within a realm of quantitative examination using the system. Now additional apparatus for the study of this etalon-fringe dynamics are still under preparation.

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