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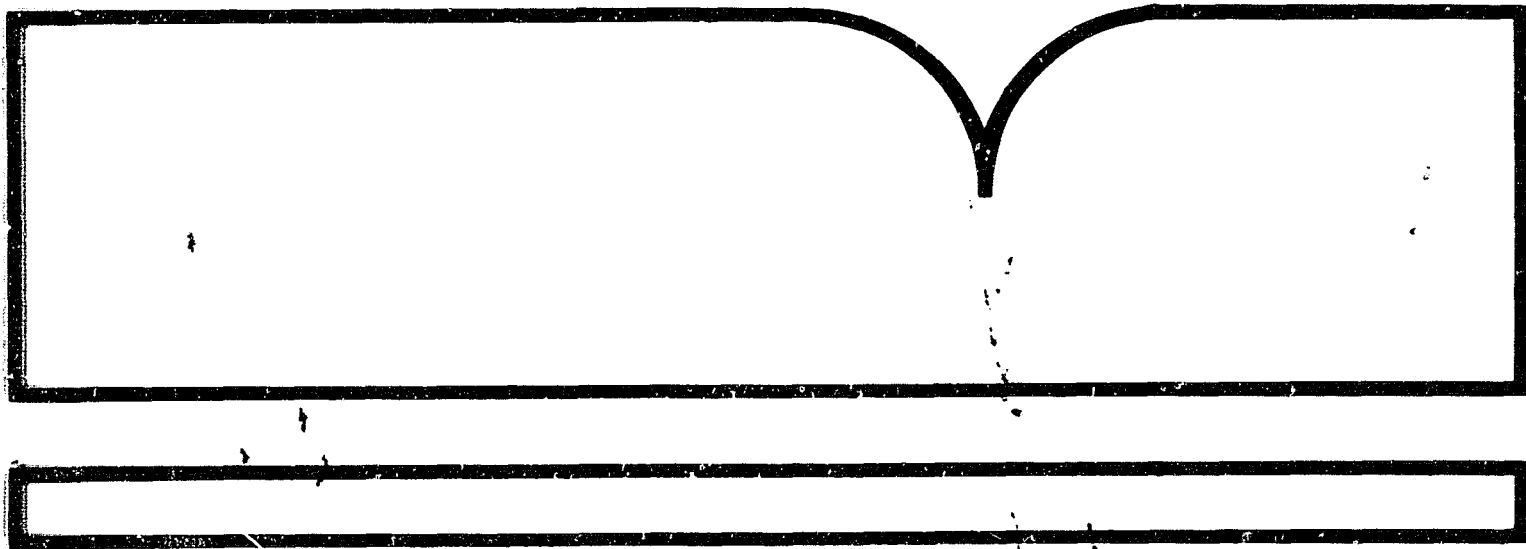
Laser System for Natural Gas Detection  
Phase I: Laboratory Feasibility Studies

Jet Propulsion Lab.  
Pasadena, CA

Prepared for

Gas Research Inst.  
Chicago, IL

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| <p>This project demonstrated the feasibility of using laser remote sensing technology as a tool for leak survey work in natural gas distribution systems. A laboratory device was assembled using a pair of helium neon (HeNe) lasers to measure methane. One HeNe laser emits radiation at a wavelength of 3.3922 <math>\mu\text{m}</math>, which corresponds to a strong absorption feature of methane, while the other emits radiation at a wavelength of 3.3911 <math>\mu\text{m}</math>, which corresponds to a weak absorption by methane. As a particular area is scanned for leaks, the laser is pointed at convenient topographic targets within its operating range, about 25 m. A portion of the backscattered radiation is collected by a receiver and focused onto an indium antimonide (InSb) photodetector, cooled to 77 K. Methane concentrations were determined from the differential absorption at the two wavelengths for the backscattered radiation.</p> <p>Laboratory tests were performed to define useful range, detection limits, spectral interferences, and variations in target reflectance. A field-transportable model was constructed and tested at a sanitary landfill and at several sites along an underground gas distribution system where methane was venting into the atmosphere. At a range of 13 m, the detection limit for methane was estimated to be 3 ppm for a 1-m pathlength.</p> |  |                                    |  |  |
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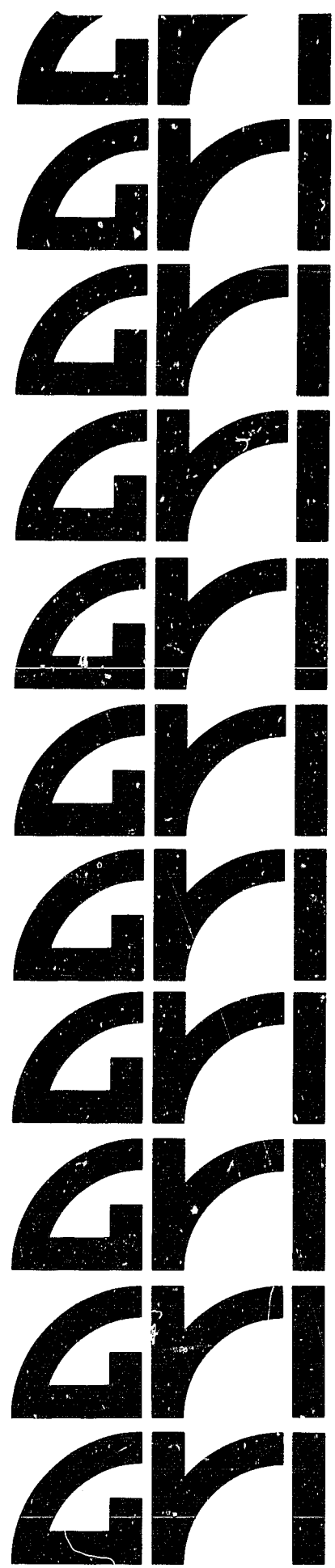
**Phase I**

**Laboratory Feasibility Studies**

**Annual Report  
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**Gas Research Institute  
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Laser System for Natural Gas Detection

Phase I - Laboratory Feasibility Studies

Annual Report

July 21, 1982

Prepared by: William B. Grant  
E. David Hinkley  
Jet Propulsion Laboratory  
California Institute of Technology  
Mail Stop 183-401  
4800 Oak Grove Drive  
Pasadena, California 91109  
(213) 354-2750

for: Dr. L. L. Altpeter  
Gas Research Institute  
8600 West Bryn Mawr  
Chicago, Illinois 60631

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Phase I - Laboratory Feasibility Studies

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Objective To determine the best lasers and measurement techniques for portable and/or transportable systems for the remote detection of leaks from natural gas pipelines.

Technical Perspective Natural gas distribution companies must survey the delivery pipeline system for leaks on a regular basis not to exceed five years, and repair the major leaks found. Currently, persons walk over the pipelines using flame ionization detectors to search for the leaks. It is expected that laser remote sensors will speed up the survey process, resulting in lower costs to the companies and their customers.

Results The feasibility of laser remote sensing technology as a tool for leak survey work in gas distribution systems was successfully demonstrated. A laboratory device was constructed using a pair of helium neon (HeNe) lasers to measure methane in leaks of natural gas.

Laboratory tests were performed to define useful range, detection limits, spectral interferences, and variations in target reflectance. A field-transportable model was built and taken into the field where sources of methane venting into the atmosphere were measured at a sanitary landfill and at several points along a gas distribution system. At a range of 13 m, the detection limit for methane (above its global concentration of 1.6 ppm) was estimated to be 3 ppm in a gas plume with a path length of 1 m (or 30 ppm for a 10 cm path-length.)

A survey of lasers for the concomitant measurement of ethane identified one or more candidates, but more development work would be required before a field-usable system could be designed around one.

Technical Approach The concept of the first generation remote sensor for leaking natural gas was based on the use of the HeNe laser. It was selected for several reasons: it possesses a strong emission line at a wavelength which corresponds to a strong absorption line of methane in the infrared spectral region; it is a low-cost, reliable device which is eye-safe and easy to operate; and it operates in a region where background radiation is minimal. Methane in leaking natural gas was detected by using the differential absorption lidar (DIAL) technique.

In the field, the laser was directed at convenient topographic targets, which provided the backscattered radiation to a collecting mirror mounted coaxially with the laser. At the focus of the mirror was an indium antimonide (InSb) detector cooled to 77 K. The presence of a natural gas leak was detected when there was a much stronger absorption at the signal wavelength (3.3922 micron) than at the reference wavelength. The signal was proportional to the concentration-pathlength product of the methane in the gas plume.

Initially, a breadboard model of the remote sensor was assembled on an optical table in the laboratory. The device was tested for stability, signal-to-noise ratio, variations in target reflectance, range of operation, and spectral interferences. After successful performance in the laboratory, a field-transportable system was designed and assembled in which the optical components and electronics were carried on a cart with 10" dia. pneumatic wheels. A convenient source of venting methane was found at a nearby sanitary landfill, and arrangements were also made with a local gas distribution company to test the remote sensor with previously identified natural gas leaks. Methane leaks at a range from 3 to 25 m were measured.

#### Project Implications

JPL has developed a proof-of-concept device which has demonstrated that remote sensing technology can be applied to natural gas leak detection in the gas distribution system. The remote sensing approach represents an improvement in the state-of-the-art because of the greater speed and efficiency with which the distribution system can be surveyed.

The goals of the support by GRI are to develop of a man-portable remote-sensing system, applicable to the survey of service lines, and a van-portable system which can be used to survey street mains. GRI intends to continue the support of JPL in this area in efforts which will lead to the development of a prototype device.

GRI Project Manager  
Dr. L. L. Altpeter



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## ABSTRACT

This project demonstrated successfully the feasibility of laser remote sensing technology as a tool for leak survey work in natural gas distribution systems. A laboratory device was assembled using a pair of helium neon (HeNe) lasers to measure methane, which typically constitutes 90% or more of natural gas. One HeNe laser emits radiation at a wavelength of 3.3922 microns, which corresponds to a strong absorption feature of methane. The other emits radiation at 3.3911 microns, which corresponds to a weak absorption by methane, and serves as a reference wavelength.

Methane concentration was determined using the differential absorption lidar (DIAL) technique. As a particular area is scanned for leaks, the laser is pointed at convenient topographic targets within its operating range, about 25 m. A portion of the backscattered radiation is collected by a receiver mirror mounted coaxially with the laser and focused onto an indium antimonide (InSb) photodetector, cooled to 77 K. The presence of methane is detected when there is a much stronger absorption at the signal wavelength (3.3922 microns) than at the reference wavelength. The concentration-pathlength of methane is given by the log ratio of the signals at the two wavelengths.

Laboratory tests were performed to define useful range, detection limits, spectral interferences, and variations in target reflectance. A field-transportable model was constructed and tested at a sanitary landfill and at several sites along an underground gas distribution system where methane was venting into the atmosphere. At a range of 13 m, the detection limit for methane (above its global concentration of 1.6 ppm) was estimated to be 3 ppm for a 1-m pathlength (or 30 ppm for a 10-cm pathlength). The system also performed well in the presence of high concentration-pathlengths.

## I. INTRODUCTION

The initial goal of this project was to perform a feasibility study on the use of lasers for the remote detection of natural gas. While the primary interest was in remote detection of gas leaks in processing, storage, transportation, and distribution systems, there was also an interest in using an airborne version of the system to locate underground deposits of natural gas.

An important criterion was that the laser system be relatively inexpensive and simple to operate. This condition was met through use of conventional helium-neon (HeNe) infrared lasers which lase at  $3.39 \mu\text{m}$ , where methane ( $\text{CH}_4$ ) has a strong absorption feature. Laboratory feasibility studies were conducted with HeNe lasers, followed by the assembly and demonstration of a HeNe laser system for field measurements of methane. A lightweight, portable HeNe system can be designed based on these studies.

A detection capability for ethane was desired as well, since ethane is a minor constituent of natural gas. Remote detection of ethane requires a laser operating at one or more wavelengths generated by tunable lasers. Evaluations were made of both available lasers and those under development, but none were found suitable.

## II. BACKGROUND

### A. Spectroscopy of Methane

The spectrum of methane ( $\text{CH}_4$ ) measured in low resolution shows that  $\text{CH}_4$  has two spectral regions with strong features from  $1250$  to  $1370 \text{ cm}^{-1}$  ( $8.0 \mu\text{m}$  to  $7.3 \mu\text{m}$ )<sup>(1)</sup> and  $2900$  to  $3100 \text{ cm}^{-1}$  ( $3.5$  to  $3.2 \mu\text{m}$ ).<sup>(1,2)</sup> In addition, there is a weaker overtone band near<sup>(3)</sup>  $6020 \text{ cm}^{-1}$  ( $1.66 \mu\text{m}$ ). While the strengths of the first two bands are similar, lasers are better developed in the  $3\text{-}\mu\text{m}$  region. Also, solar radiation decreases rapidly beyond  $3 \mu\text{m}$ , and the Earth's blackbody radiation increases from  $4$  to  $10 \mu\text{m}$ , so the  $3\text{-}\mu\text{m}$  region has the least interference from background radiation.

### B. Spectroscopy of Ethane

Ethane has three strong absorption bands centered at  $3.4$ ,  $6.8$  and  $12 \mu\text{m}$ .<sup>(1)</sup> The  $3.4\text{-}\mu\text{m}$  band is the strongest. The  $6.8\text{-}\mu\text{m}$  band occurs in a region where water vapor has many strong absorption lines. The  $12\text{-}\mu\text{m}$  band is much weaker and is in a region where few lasers operate well. The  $3.4\text{-}\mu\text{m}$  region is the best region to try to measure ethane because the absorption lines are strongest, spectral interference is small (except for other hydrocarbons), and detector noise is relatively low.

A high resolution spectrum of ethane was obtained.<sup>(4)</sup> It was taken at  $3.9$  torr, with a resolution of  $0.02 \text{ cm}^{-1}$ . At standard atmospheric pressure, the lines will broaden by about  $0.1 \text{ cm}^{-1}$ . The spectrum shows a number of strong absorption lines in the  $3.307\text{-}$  to  $3.385\text{-}\mu\text{m}$  spectral region ( $2954$  to  $3024 \text{ cm}^{-1}$ ). The strongest lines approach a strength of  $25 \text{ atm}^{-1}\text{cm}^{-1}$ , about three times that employed for methane with HeNe lasers.

### C. Differential Absorption Technique

Using lasers, the most sensitive way to measure gases in the lower atmosphere remotely is with the differential absorption technique.<sup>(5-7)</sup> Here, one laser wavelength, at which the gas of interest absorbs strongly, is used to detect that gas; a second wavelength, at which the gas absorbs weakly, is used for reference purposes to eliminate other variables. A ratio of the signals at the two wavelengths, suitably corrected for transmitted power and target distance, is used to determine gas concentrations using Beer's law, for the case of a plume in the presence of a background or ambient concentration:

$$c\ell + CR = \frac{-1}{2\Delta\alpha} \left\{ \ln\left(\frac{I}{I_0}\right) - \ln\left(\frac{I'}{I_0'}\right) \right\} \quad \text{Eq. 1}$$

where  $c$  is the concentration of gas in the plume (atm or, equivalently, ppm)  
 $\ell$  is the diameter of the plume (cm)  
 $C$  is the ambient concentration of the gas (atm)  
 $R$  is the distance to the target (cm)  
 $\Delta\alpha$  is the differential absorption coefficient (atm<sup>-1</sup> cm<sup>-1</sup>)  
 $I$  is the received power for the "on" ("off") wavelength (W)  
 $I_0$  is the transmitted power for the "on" ("off") wavelength (W).

In order to use Equation 1 directly with one radiation detector, the beams can be chopped 90° out of phase and two phase-synchronous detectors used to analyze the signals. Alternatively, the beams can be chopped 180° out of phase and blocked successively and one phase-synchronous detector used. For both of these methods, ratios of the signals are formed to determine the methane concentration-pathlength product.

Another way to use the system is to chop the beams 180° out of phase and send both signals to one phase-synchronous detector and take the difference between the two signals. The signal at the "on" wavelength has the form:

$$I = \frac{I_0 A \rho n f(\theta) \exp(-2\alpha(c\ell + CR))}{\pi R^2} \quad \text{Eq. 2}$$

where  $A$  is the receiver area (cm<sup>2</sup>)  
 $\rho$  is the reflectance of the target  
 $f(\theta)$  is a function that depends on the surface roughness of the target and the angle of incidence of the lidar beam  
 $n$  is the receiver efficiency.

The signal at the "off" wavelength can be formed from Eq. 2 by adding primes to the variables that change with wavelength. The difference signal is then:

$$I - I' = \frac{A n p f(\theta) \exp(-2\alpha(c\ell + CR)) - \rho' \exp(-2\alpha'(c\ell + CR))}{\pi R^2} \quad \text{Eq. 3}$$

For the case where  $\rho = \rho'$ , it becomes

$$I - I' = \frac{A n p f(\theta) \exp(-2\alpha(c\ell + CR)) - \exp(-2\alpha'(c\ell + CR))}{\pi R^2} \quad \text{Eq. 4}$$

And for the case where  $2\alpha(c\ell + CR) \ll 1$ , it becomes

$$I - I' = \frac{-A n p \Delta \alpha f(\theta)(c\ell + CR)}{\pi R^2} \quad \text{Eq. 5}$$

which is directly proportional to the concentration and thus represents a potentially very useful technique for detecting small leaks. However, since the ratio of  $\rho$  to  $\rho'$  may vary with the target materials, this approach could lead to scanning-system problems (see Sec. III G). Note that this approximation is valid for concentration-pathlengths less than about 100 ppm-m, for which the error is less than 5%. Also note that the electronics becomes simpler in this case because only one and not two signal processing channels are required, since a difference, rather than a ratio, is measured.

### III. LABORATORY MEASUREMENTS

#### A. Choice of Laser for CH<sub>4</sub>

In the 3- $\mu\text{m}$  band, the HeNe laser at  $2947.9 \text{ cm}^{-1}$  (3.3922- $\mu\text{m}$  vacuum wavelength or 3.3913  $\mu\text{m}$  in air) has a strong overlap with a methane line (absorption coefficient  $(\alpha) \approx 8.8 \text{ cm}^{-1} \text{atm}^{-1}$ ).<sup>(2)</sup> In addition, the HeNe laser can be made to lase at  $2948.9 \text{ cm}^{-1}$  ( $\lambda$  vacuum = 3.3911  $\mu\text{m}$ ) with the addition of an intracavity cell containing CH<sub>4</sub> to quench the dominant line<sup>(8)</sup> at  $2947.9 \text{ cm}^{-1}$ . The HeNe laser has been used in several laboratory<sup>(9,10)</sup> and field<sup>(11)</sup> demonstrations of the measurement of CH<sub>4</sub>. Other lasers that have been used for remote measurement of CH<sub>4</sub> on weaker lines include the OPO laser,<sup>(3)</sup> the DF laser,<sup>(12)</sup> the Er: YAG laser,<sup>(13)</sup> and the diode laser.<sup>(14)</sup>

#### B. Preliminary Laboratory Tests

After the HeNe laser was chosen as the best laser for use in a simple system for the remote measurement of natural gas, a breadboard version was assembled on an optical table in the laboratory. Then a series of tests were performed to evaluate the likelihood that a field system could be constructed to meet user requirements.

### C. Laboratory Test System

A block diagram of the system used in the laboratory is shown in Figure 1, and the components and their parameters are listed in Table I. The HeNe laser beams for the "on" and "off" wavelengths are made parallel and nearly colinear by means of a pair of mirrors. They are separated just enough so that they can be chopped 180° out of phase by the small-aperture (5 mm) chopper that operates at 2 kHz. Two additional mirrors are used to make both beams coaxial with the receiver axis. Backscattered radiation from a topographic target is collected by a 37-cm diameter receiver mirror and directed through an optical narrow-band filter to an InSb detector. The filter reduces background radiation by a factor of four. The detected radiation signal is amplified and processed by a lock-in (phase -synchronous) detector.

The helium-neon (HeNe) lasers operate continuously (CW) and emit a beam with a divergence of about 1 mrad (expands to 1 m in 1 km). The Spectra Physics Model 120 laser emits 1.6 mW at 3.3922  $\mu\text{m}$  ( $\alpha_{\text{CH}_4} \approx 8.8 \text{ atm}^{-1}\text{cm}^{-1}$ ); the Spectra Physics Model 124B laser emits 2.2 mW at 3.3911  $\mu\text{m}$  ( $\alpha_{\text{CH}_4} \approx 0.3 \text{ cm}^{-1}\text{atm}^{-1}$ ). The chopper wheel is operated at 2 kHz, which is high enough to reduce much of the mechanical (1/f) noise occurring at lower frequencies.

### D. Measurement of System Signal and Noise

The signal power measured by a laser system is given by Equation 2. For the laboratory system, measurements were made assuming the following values of the parameters:

$$\begin{aligned} I_0 &= 1.5 \text{ mW} \\ \rho &= 0.07 \\ A &= 0.095 \text{ m}^2 \\ \eta &= 0.2 \\ R &= 15 \text{ m} \\ \alpha &= 8.8 \text{ cm}^{-1}\text{atm}^{-1} \\ C &= 1.6 \cdot 10^{-6} \text{ atm} \\ \text{and } c\lambda &= 0. \end{aligned}$$

Putting these values in Eq. 2 yields a value of  $2.7 \cdot 10^{-9}$  W.

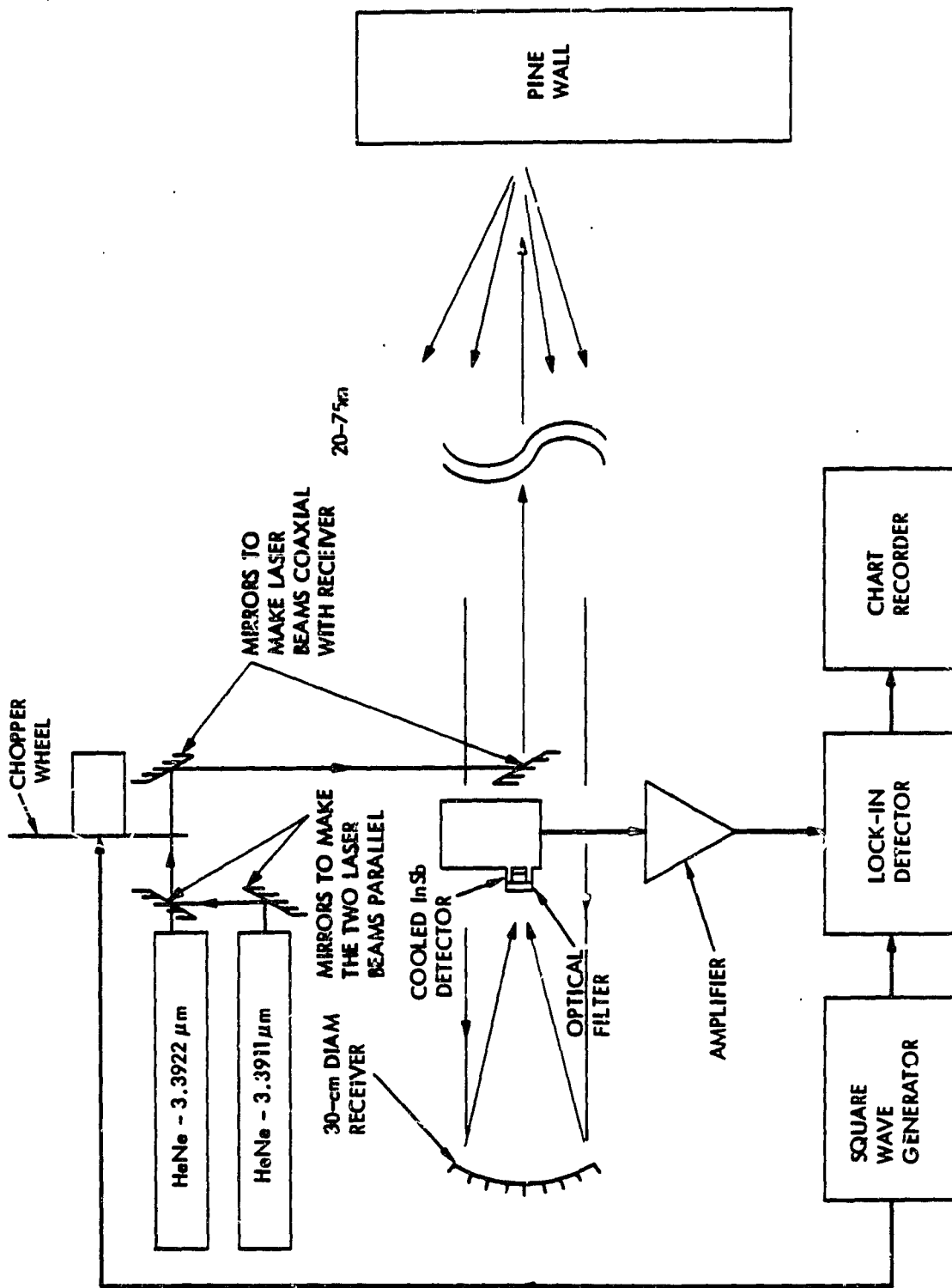


Figure 1. Block diagram of the HeNe Laser System for Remote Measurement of Methane

TABLE I

Characteristics of the HeNe Laser System

Transmitter

HeNe Laser (2 ea)  
Input power 50 - 100 W  
Emitted power 1.5 mW  
Beam divergence 1 mrad  
Chopper frequency 2 kHz  
Wavelength - "on" 3.3922  $\mu\text{m}$  (vacuum)  
Wavelength - "off" 3.3911  $\mu\text{m}$  (vacuum)  
(intracavity cell containing  $\text{CH}_4$  used)

Receiver

Mirror  
Diameter 14.5" (36.8 cm)  
Focal length 34.5" (87.6 cm)  
Surface spherical  
Coating aluminum with  $\text{SiO}_2$  overcoat

Detector  
Material InSb  
 $D^*$   $\sim 10^{11}$   $\text{cm Hz}^{1/2} \text{W}^{-1}$   
Diameter 2 mm  
Temperature 77 K

Filter  
Bandwidth 4.2%  
Peak transmission 70%  
Temperature 77 K

System optical efficiency = 0.2

Signal processing electronics  
Phase-sensitive (lock-in) detectors (2 ea)  
(sensitive from 1  $\mu\text{V}$  to 500 mV)  
Dual-channel chart recorder

System Cart  
Table top - Kevlar  
Filler - honeycomb aluminum  
Frame - angle aluminum  
Wheels - 10" diameter pneumatic  
Height - 1 m

Electrical power generator  
1500 W AC, 120 V (500 W required)



The detector noise equivalent power (NEP) is given as

$$\text{NEP} = \frac{A_d^{1/2} \tau^{-1/2}}{D^*} \text{ (watts)} \quad \text{Eq. 6}$$

where  $A_d$  is the area of the detector ( $\text{cm}^2$ )  
 $\tau$  is the integration time  
 and  $D^*$  is the detectivity of the detector ( $\text{cm Hz}^{1/2} \text{ W}^{-1}$ )

For the laboratory system,  $A_d = 0.03 \text{ cm}^2$ ,  $\tau = 1 \text{ sec}$  and  $D^* = 10^{11} \text{ cm Hz}^{1/2} \text{ W}^{-1}$ . Putting these values in Eq. 6 yields a detector NEP of  $3.10^{-13} \text{ W}$ .

With the narrow band filter (FWHM = 4.2%), solar and thermal background radiation also contribute to the system noise. At  $3.4 \mu\text{m}$ , the combined background radiation is about  $10^{-4} \text{ W}/(\text{cm}^2 \text{ sr } \mu\text{m})$ . For our system, this calculates to be  $1.33 \cdot 10^{-13} \text{ W}$ .

The signal-to-noise ratio (S/N) for a 1-sec. integration time can be estimated from the signal, calculated using Eq. 2 with a target distance of 12 m and the combined noise from the detector and background radiation (Eq. 6). The calculated ratio is 6,200. The value measured in the laboratory for a pine target is 1330. The discrepancy is a factor of 4.7.

A signal-to-noise ratio of 100 means that there is 1% measurement error using Eq. 2. This would translate, in the case of methane, to an uncertainty in the concentration-pathlength of 6 ppm-m (6 ppm in 1 m) using Eq. 1. (This is twice the measurement error for one measurement, since four power measurements have to be made for one concentration measurement.)

#### E. Target Reflectance Variation

A variety of targets were used to check the signal as a function of material and reflectivity. Vegetation gave about the lowest signal, asphalt an intermediate signal, wood and gravel a fairly high signal, and crumpled aluminum a signal 20 times that for asphalt. The reflectivities of most natural non-vegetation targets were within a factor of 3 of each other.

#### F. Effect of Chopper Frequency

The chopper frequency was varied from 360 to 2400 Hz, and the system signal-to-noise ratio (S/N) was determined for each setting for one laser wavelength. The laser was pointed at the pine wall 20 m away, and an integration time of 0.1 sec was used. The S/N increased from 11 at 360 Hz to 22 at 2400 Hz in a monotonic fashion. The detector curve of detectivity versus chopping frequency supplied by the manufacturer indicates an improvement as the frequency is increased to about 1 kHz. The extra improvement could come from a further reduction in sensitivity to mechanical vibrations, including that of the chopper itself, which was mounted on rubber feet.

#### G. Differential Spectral Reflectance

Differential spectral reflectance changes, i.e., changes in the relative reflectance at two wavelengths, can be a serious source of noise for laser systems that use topographic targets.<sup>(15)</sup> The HeNe laser wavelengths chosen

during this study are approximately  $1 \text{ cm}^{-1}$  apart. Between 3- and 4- $\mu\text{m}$ , some targets, such as paint, may have a factor of four change in reflectance, while others may have no change. (16,17) In other words, there can be up to a 400% change in reflectance in a region containing  $833 \text{ cm}^{-1}$ . This corresponds to a maximum of about 0.5% for the  $1 \text{ cm}^{-1}$  separation of the two HeNe laser lines used for methane detection. Usually, however, the effect would be considerably less, probably about 0.05%, which corresponds to a methane measurement error of 0.3 ppm-m.

H. Effect of Changing Target Distance

The image plane of a receiver mirror varies with distance to the object by the relation:

$$\frac{1}{f} = \frac{1}{i} + \frac{1}{o} \quad \text{Eq. 7}$$

where  $f$  is the focal length  
 $i$  is the mirror to image distance, and  
 $o$  is the mirror to object (target) distance.

For a 30-cm-diameter spherical mirror, with a focal length of 105 cm, the change in the image plane is quite noticeable at short receiver-to-target distances. Some values are given here:

Table II. Variation of Image Plane with Target Distance for a 105-cm-Focal-Length Receiver

| Target Distance (o) | Image Plane (i) |
|---------------------|-----------------|
| (m)                 | (cm)            |
| 10                  | 117.3           |
| 30                  | 108.8           |
| 60                  | 106.9           |
| 100                 | 106.1           |
| $\infty$            | 105             |

This variation has the effect of reducing the detected signal for target distances other than that for which the detector is set. Essentially, the image is blurred over a larger diameter as the range changes. There is some mitigation of this effect by the  $1/R^2$  increase in signal from a topographic target as the range (R) is decreased. An example of how this operates is shown in Figure 2, for detector diameters of 1, 2 and 3 mm, a setting range of 60 m and 30 m, and a laser divergence of 1 mrad. Note that the effect is less pronounced for the larger detectors. The noise of a detector due to blackbody radiation normally increases linearly with the diameter. For the case shown, a 2-mm diameter detector (such as the one we have) would be superior to a 1-mm diameter detector for the range 44-84 m, while a 3-mm diameter detector would be better if an extended range were to be used without changing the detector position. If a limited set of ranges were to be used, it would be better to change the detector position than go to a larger detector. Laboratory measurements confirmed the general shape of the return signal with distance.

### I. Spectral Interference by Other Hydrocarbons

Many hydrocarbons have strong, broad absorption bands somewhere in the spectral region 3.2 to 3.5  $\mu\text{m}$ . This arises from the C-H stretch vibrational mode. Using the low resolution spectra available, (e.g., in the Sadtler Spectral Atlas (1)) it is difficult to tell whether particular hydrocarbons would interfere with a measurement of methane. In order to interfere, the gas would have to have a differential absorption for the two wavelengths chosen for the methane measurement. By choosing two lines close together as we have with the HeNe laser, the interference should be minimized.

In order to assess this effect, the absorption coefficients of several hydrocarbons were measured in the laboratory. To do this, an 8-liter metal box with quartz windows separated by 30 cm and a fan for circulating the gas-air mixture was placed in the path of the laser beams. A syringe was used to inject 10 to 25 ml of gas into the box, giving concentrations on the order of 1000 to 2500 ppmv. The results of the measurement are given in Table III. Note that some gases, such as butane and propylene, have small differential absorption coefficients at the two HeNe wavelengths (acetylene and benzene have no absorption at 3.39  $\mu\text{m}$ ), while others, such as ethane, isobutane and propane, have differential absorption coefficients that are large enough to cause noticeable interference - or to allow these gases themselves to be detected near their source, e.g., leaking propane near a propane storage tank. Note, too, that the sign of the differential absorption coefficient changes for some gases. Thus, in a mixture of hydrocarbons, such as gasoline, the combined differential absorption-coefficient might be quite small. However, automobile exhaust, which can contain up to 10%  $\text{CH}_4$  (18) of the total hydrocarbon, which is only a few per cent of the gaseous emissions, might give a measurable signal.

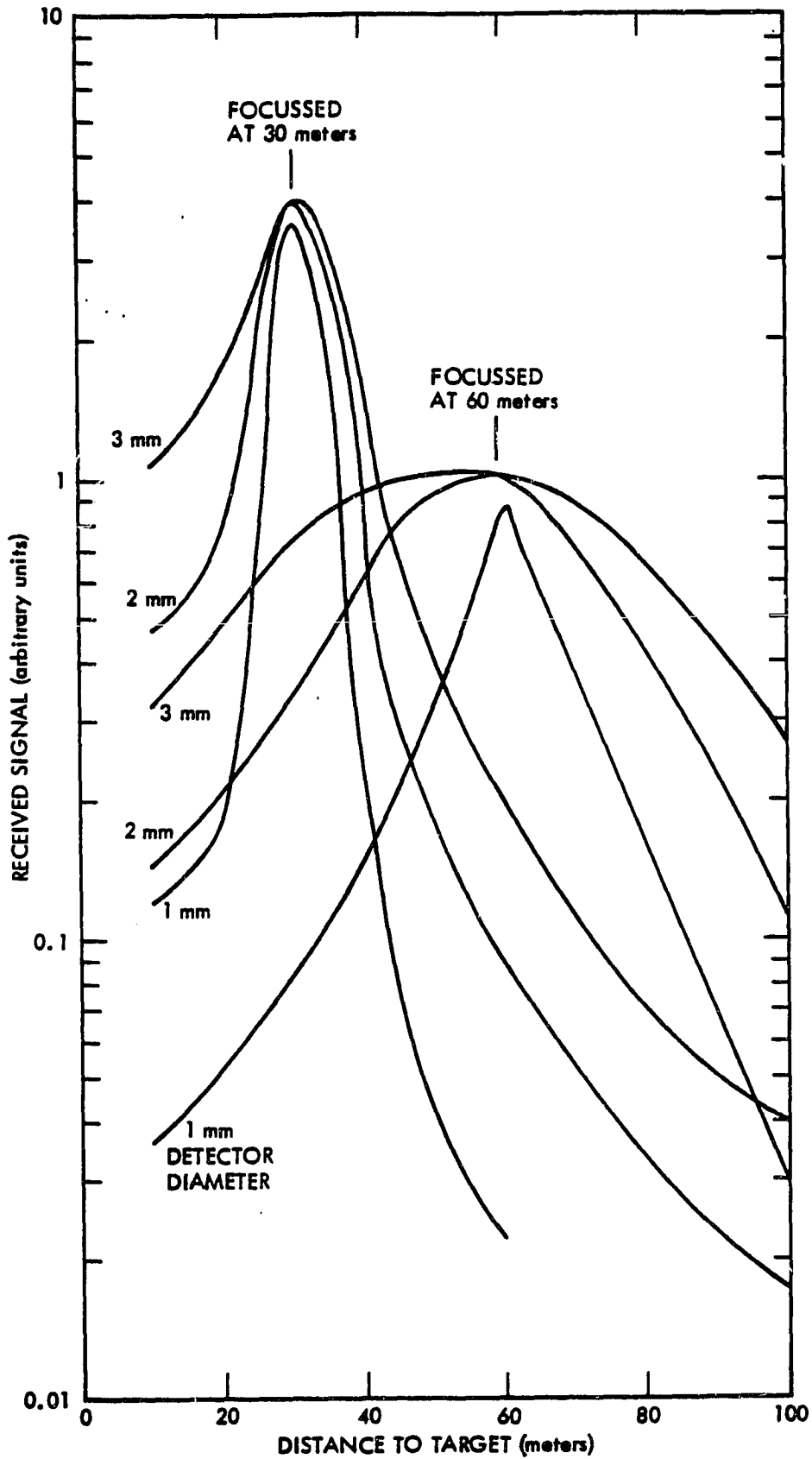


Figure 2. Calculated Signal Versus Distance to Target for Two Focal Positions of Three Detectors.

Table III. Hydrocarbon Absorption Coefficients at HeNe Wavelengths

| Gas       | $\alpha_{3.3922}$<br>( $\text{atm}^{-1}\text{cm}^{-1}$ ) | $\alpha_{3.3911}$<br>( $\text{atm}^{-1}\text{cm}^{-1}$ ) | $\Delta\alpha$<br>( $\text{atm}^{-1}\text{cm}^{-1}$ ) |
|-----------|--|--|---|
| Methane   | 8.8(2)   | $0.8 \pm 0.3$  | $+8.0 \pm 0.3$  |
| Butane    | $10.9 \pm 0.4$   | $11.0 \pm 0.4$   | $-0.1 \pm 0.6$  |
| Ethane    | $3.69 \pm 0.19$  | $2.69 \pm 0.08$  | $+1.0 \pm 0.2$  |
| Isobutane | $11.1 \pm 0.2$   | $13.0 \pm 0.4$   | $-1.9 \pm 0.4$  |
| Propane   | $8.27 \pm 0.04$  | $7.41 \pm 0.03$  | $+0.86 \pm 0.06$                                      |
| Propylene | $2.23 \pm 0.06$  | $2.40 \pm 0.02$  | $-0.17 \pm 0.06$                                      |

J. Laser Stability

One instrument, a Spectra Physics Model 120 HeNe laser, had a fraction of a percent power drift in a period of a few minutes. The other, a Spectra Physics Model 124B HeNe laser, had a power instability in the form of a sine wave with an amplitude of  $\pm 4\%$  and a period of about one minute during each first day of operation. After a day or two, it became rather quiet, with less than 1% per minute drift. The cause of the problem appears to be related to thermal expansion of the aluminum channel supporting the optical cavity.

K. Evaluation

The results of this portion of the work indicated that an adequate laser remote sensing system could be built from inexpensive, commercially available components, and that it could be used in the field with topographic targets to locate leaks from underground natural gas pipelines.

IV. FIELD SYSTEM

A. System Description

A field version of the laboratory laser remote sensor of methane was designed and assembled. A photograph of this system is shown in Figure 3. The optical components are on one level of the cart, while the electronics are primarily on the other level. Ten-inch-diameter pneumatic wheels help isolate the system from vibrations as it is transported. The system is self-sufficient in that a gasoline-powered electrical generator can be used to supply electrical power.

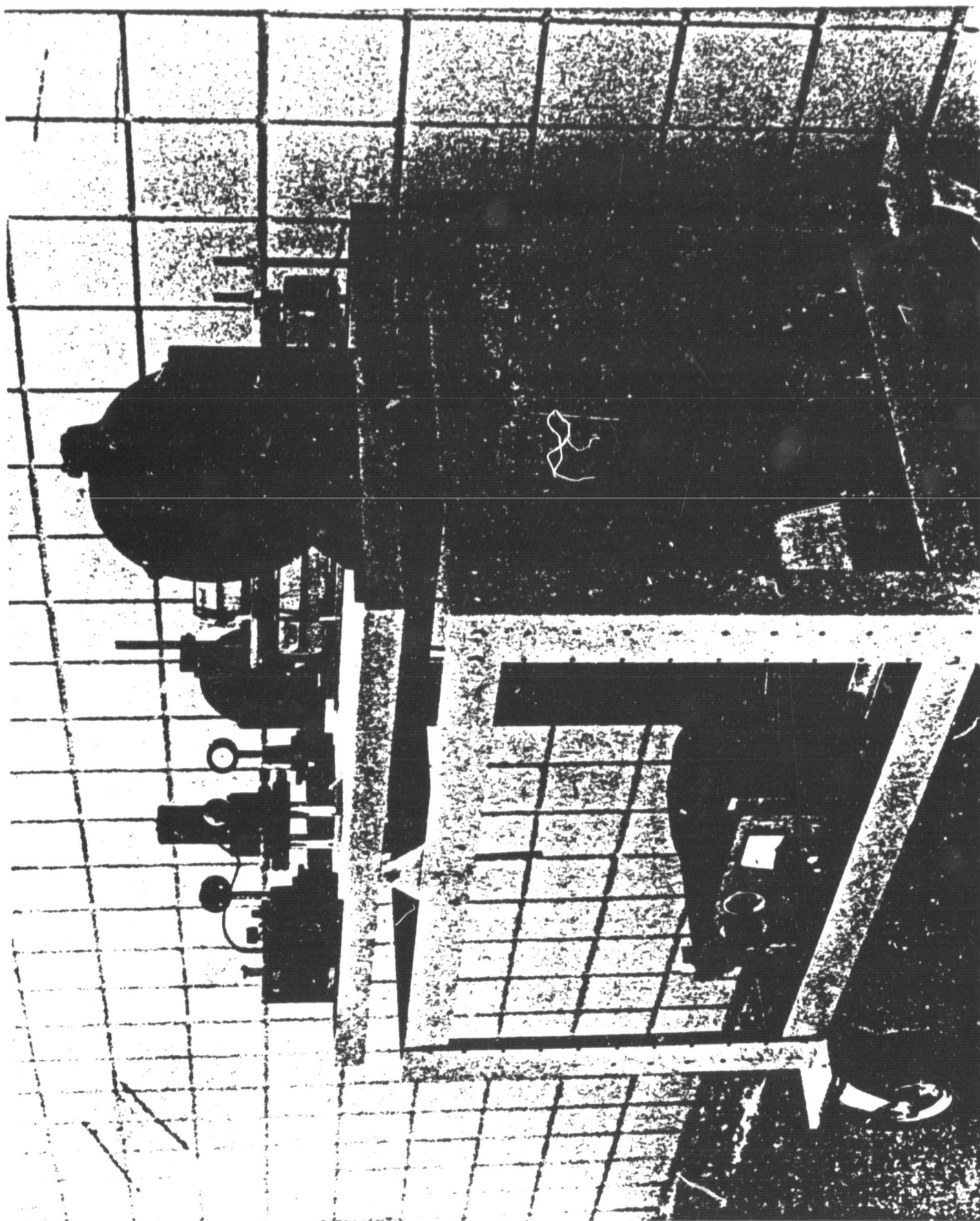


Figure 3. Photograph of HeNe Laser System.

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best available copy.



The optical components are mounted on a Kevlar-coated, aluminum honeycomb table. Kevlar has a very low thermal expansion coefficient. This table can be tilted  $\pm 10^\circ$  and can be interchanged with the lower shelf.

Many of the components used in the breadboard laboratory version were incorporated into the field system. One improvement was that a rigid 1.5-inch-diameter pole with micrometer-screw adjust was used to mount the two mirrors which bring the two laser beams into near coincidence with each other. The receiver mirror was replaced by a 14.5-inch- (37-cm) diameter mirror with tapered edges to reduce the weight to 16 pounds, a 2-inch (5-cm) hole in the center, and a focal length of 34.5 inches (88 cm).

The electronics was augmented by the addition of a second phase-synchronous detector so that signals from each of the lasers could be measured independently. A 1500-W electrical generator was purchased to provide power for the system. This later proved to be larger than necessary, since the system draws only 300 W.

The table with the optics and electronics weighs about 250 - 300 pounds. Again, this is much larger than would be required for a well-designed field system.

#### B. Laboratory Tests

Several tests were performed with the system at JPL, pointing it out of the laboratory down the asphalt road leading to the east.

In one set of tests, the laboratory supply of natural gas was brought out to a point a few inches below the laser beams using tygon tubing. The system was aimed at the asphalt road about 13 m away. The time constant was set to 0.1 or 0.03 sec on the phase-synchronous detector. When the gas blew into the beams, the signals for both wavelengths decreased. This is shown in Figure 4. The signal at both wavelengths varies, as expected from Eq. 2, even though the absorption coefficients vary by a factor of 11. The exponential function varies much more rapidly for a given change in the exponent for small values of the exponent, so that the ratio of the measured signal intensities are less than a factor of 11. This time-varying behavior of escaping natural gas has been found to be the best indication of its presence.

#### C. Attempted Ethane Measurement

It was noted that when ethane is mixed with methane, as is the case with natural gas, the absorption coefficients at the two HeNe wavelengths change. For 3% ethane, 97% methane, the absorption coefficient at 3.3922  $\mu\text{m}$  changes from 8.8 to 8.65  $\text{atm}^{-1}\text{cm}^{-1}$ ; at 3.3911  $\mu\text{m}$  it changes from 0.8 to 0.86  $\text{atm}^{-1}\text{cm}^{-1}$ . If the absorption by the gas is measured at the two wavelengths and a plot is made of the absorbance at the signal wavelength  $\alpha c \ell$  (where  $\alpha$  is the absorption coefficient,  $c$  is the concentration, and  $\ell$  is the pathlength) versus the differential absorbance  $(\alpha_1 - \alpha_2)c \ell$ , a difference in the slope of 1% is expected between pure methane and the 97% methane/3% ethane mixture.

Several measurements were made with the laser system in the laboratory pointing at the asphalt road 10 m from the system to test this. For the two measurements with laboratory pipeline gas, the slopes were 1.053x and 1.018x that for pure methane (1.009x was expected). For one set of field

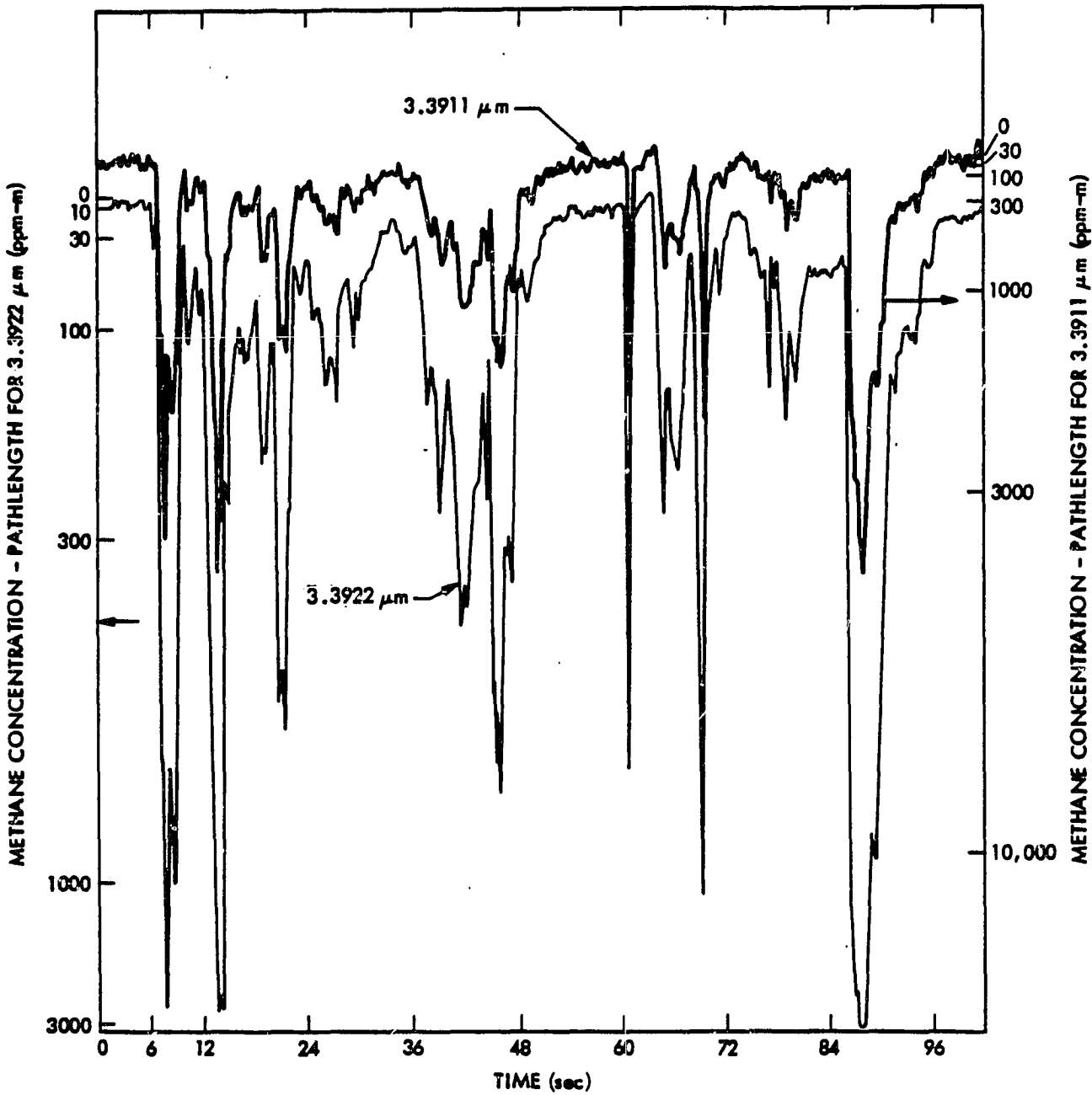


Figure 4. Time-Varying Methane Absorption Using a Topographic Target at 13 m and a 0.03 sec. Time Constant.



measurements analyzed, the ratio was 0.912x, much less than expected. These results indicate that it might be possible, but difficult and unreliable, to measure the ethane content of natural gas using the HeNe laser lines that are appropriate for methane. However, this technique requires a rather high gas concentration-pathlength product (a few hundred ppm-m) for measurement accuracy, and also seems dependent on careful system alignment.

#### D. System Signal-to-Noise Measurements

Some measurements were made with topographic targets at distances of 70 to 100 m. Concrete was used at 70 m; asphalt and concrete at 100 m. A signal strength of about 2  $\mu\text{V}$  was measured. (Noise from background radiation = 0.3  $\mu\text{V}$ .) With a few seconds of observations, the noise was reduced to about 15% of the signal. This value is equivalent to about 100 ppm-m, so that a leak of this magnitude could be detected at those ranges with this system. The signal falloff was slightly faster than the  $1/R^2$  expected using Eq. 2.

A further measurement performed at JPL was to monitor the return from a 5-inch-diameter retroreflector placed 500 m from the laser system. It was observed that the noise for either laser alone at 0.1 sec integration was about 8% of the signal amplitude, but only 3% when the difference between the returns for the two wavelengths was measured. (See Eqs. 3 - 5.) This effect is attributed to atmospheric index-of-refraction-induced variations in signal, which are correlated for the two beams passing through the same air mass. It was also determined that the signal from the retroreflector falls off approximately as  $1/R^2$ , where R is the distance. This is due to the spread of the laser beam and could be reduced by using a beam expander to reduce the divergence; but this would be difficult with two lasers.

#### E. Field Measurement of Methane - Landfill Site

The laser system was taken on the bed of a pickup truck to the Scholl Canyon landfill site, located 10 miles from JPL above the town of Eagle Rock (west of Pasadena). The area being filled is in a mountain canyon. The width is about 150 m at the east end where the measurements were made, and is somewhat greater near the center and to the west. The length of the site is about 600 m. The surface has been covered with soil which has been compacted and leveled. Filling is currently taking place along a line running north-south near the middle of the site. In the region studied with the laser system, there were numerous cracks in the soil, and the region about 15 to 35 m from the south edge had loose, rather than compacted soil.

On August 27, long-path measurements were made with a 5-inch-diameter retroreflector placed on one side of the site, the laser system still being on the pickup truck at a distance of 100 or 150 m away. The retroreflector increases the signal by a factor of 1000 or more at long ranges compared with topographic targets. The height above the ground averaged about 5 feet. The target was easy to acquire; the table top was tilted until the image of the mirror fell on the center of the detector. The final adjustments were made by moving the table and watching the signal on the lock-in detector. The beam-steering mirrors were adjusted for the first measurement only.

When measuring small amounts of methane, it suffices to measure the difference between the returns at the absorbing and reference wavelengths. As discussed in Sec. II C, this is more sensitive than the ratio technique to the presence of small quantities of methane because the electronic signal processing is done on a faster time scale which allows for the cancellation of some measurement errors. (When measuring large concentrations of methane, however, it is necessary to measure the ratio of the signals either at two distances or for the cases of some and no methane.) Since only one set of electronics was available, the signal from the reference laser was checked occasionally, and the difference between the absorbing and reference wavelength signals was measured often. Later, the reference wavelength signal was added to the difference signal to determine the absorbing wavelength signal.

The locations of the measurements and the measurement results are shown in Figure 5. At each position, measurements were made for 5 to 10 minutes with a time constant of 0.1 sec. The values reported are those above the ambient level of about 1.5 ppm. The measurements were all relative to the minimum methane measured, which was along the south side when a strong breeze came from the south, probably clearing out methane from the landfill site.

The measurement results show a high concentration over a region where there were numerous cracks in the soil, and where the technician on the site indicated that strong methane emissions had been found previously.

It should be pointed out that this measurement approach is appropriate when the site is inaccessible - if, for example, one could be only on the perimeter. The measurements can then be used to construct a two-dimensional display of the methane sources, as in X-ray tomography. (19,20)

On August 28, the laser system was aimed at the soil about 40 ft (12 m) from the receiver mirror, and the pickup truck was driven at a speed of 40 m/min (1.5 miles/hr) across the site in a north-south direction, starting at the east end. Nine traverses were made from 10:23 to 11:18 a.m. The reference wavelength signal was monitored frequently by blocking the absorbing wavelength laser. This was necessary because the distance to the soil varied along the truck path, owing to slight tilts of the surface of the landfill so that signal fall-off could be due to change in target distance, as shown in Figure 2. This affects the measurement accuracy. Occasionally, in regions where strong methane signals were found, the reference wavelength signal also decreased noticeably, indicating that other hydrocarbons might be present in the emissions as well. This would cause some measurement error, but would not preclude finding emission "hot-spots" in the landfill site.

The measurement results are shown in Figure 6. The adjacent pairs of methane profiles were made with rows of existing stakes to guide the truck. They were about 175 feet apart. Regions in the middle between the stakes were not measured. The methane profiles are placed on the figure over each 12-m swath that was measured.

The data for each adjacent pair of traverses are fairly well correlated. The wind was blowing approximately northeast. The peak concentrations in the eastern traverse of each pair are usually shifted about 10 m to the north.

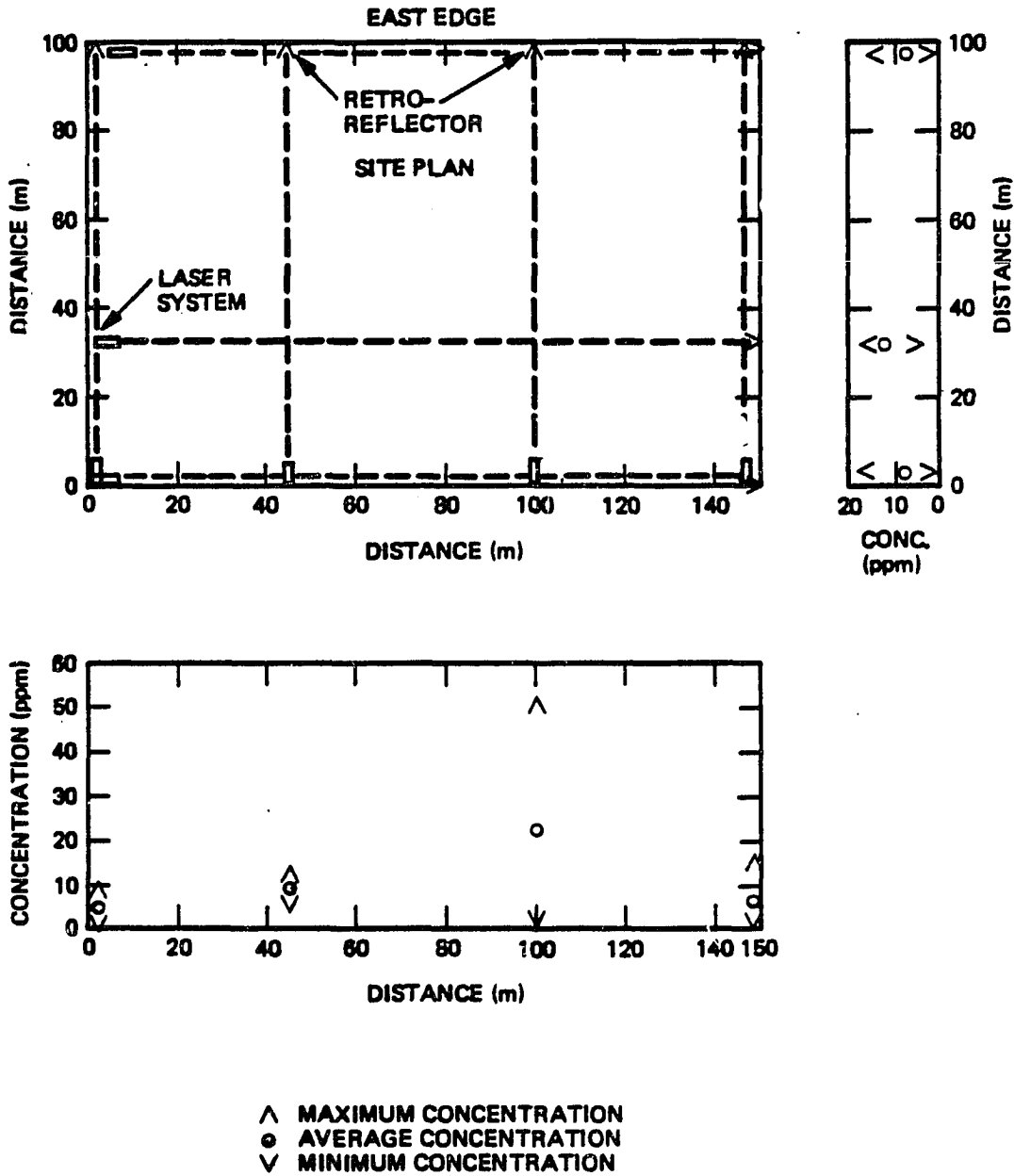


Figure 5 Measurement of Methane at the Scholl Canyon Landfill Site Using Laser Long-Path Techniques.

EAST EDGE

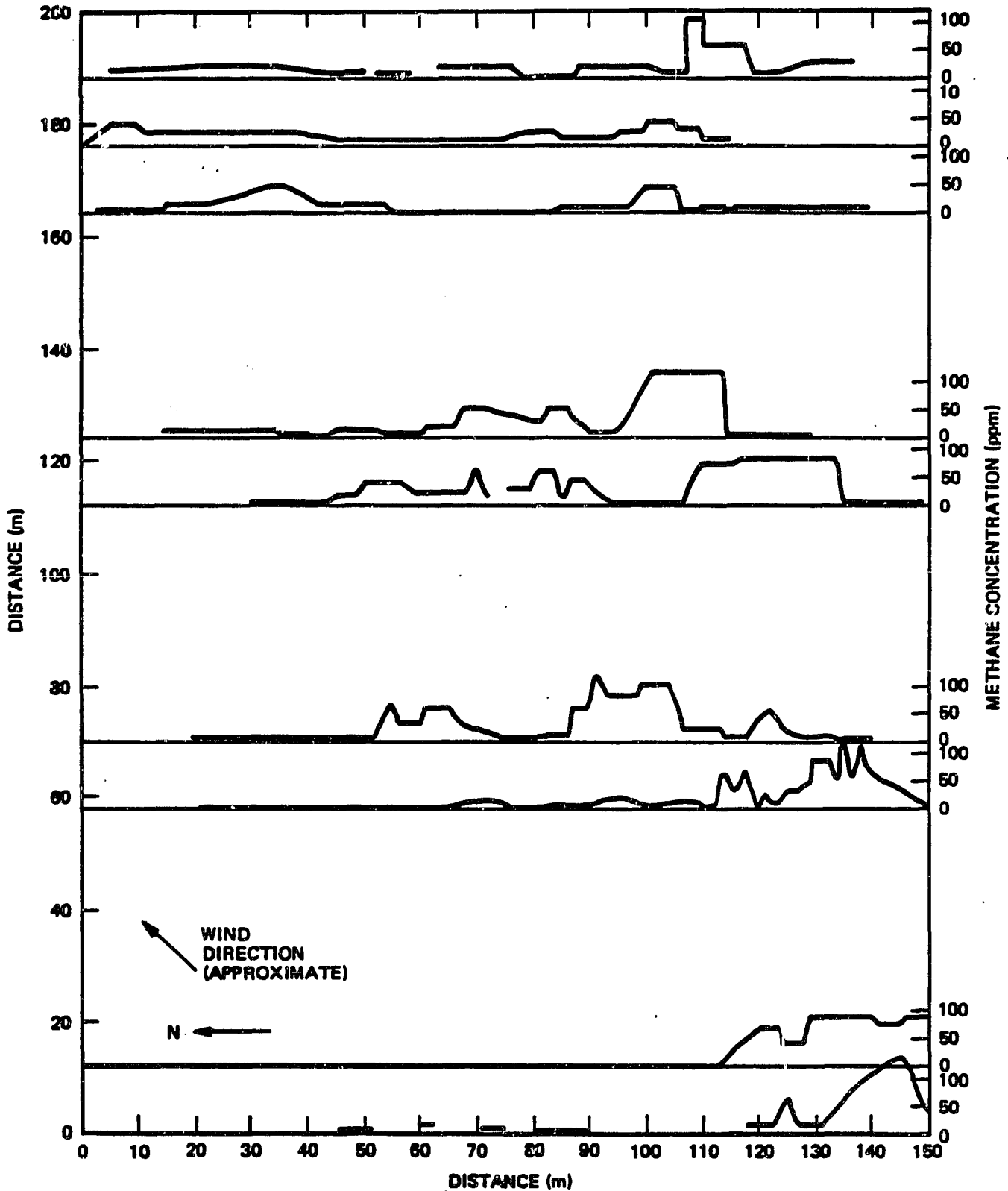


Figure 6 Methane Measured at the Scholl Canyon Landfill Site Near Eagle Rock, California, Using A Laser Remote Sensor and Backscatter From the Ground. August 28, 1981.

The lasers were about 7 feet above the ground, and the laser beam was back-scattered from the ground, so some methane emitted from a region southwest of the region covered could be measured.

The higher concentrations of methane were associated with visible cracks or a rough, uncompacted surface. The peak in Figure 5 roughly correlates with the maximum concentrations shown in Figure 6.

The entire landfill could be surveyed in about a six- to eight-hour day.

The laser system was taken to the Scholl Canyon Landfill site again on September 10. It was pointed at the ground about 14 m away. A flame ionization detector was used near the laser system to measure the methane blowing along the laser path. The system was then taken to the entry gate region above the landfill to get an ambient methane calibration. The FID measured methane concentrations up to 40 ppm. The laser system measured similar values, but usually 10 to 20% higher. Subsequently, it was learned that because of the noncoaxial transmitter arrangement, there is a range-dependent measurement error. This probably explains the discrepancy between the laser system and the FID. For a detector and laser focus at 15 m, measurements made over ranges of 11 to 24 m result in a maximum error of +20 ppm for an assumed target distance of 15 m. Even with this misalignment, reasonable data can be taken in the field if signals from both lasers are measured and it is noted whether the target distance is greater than or less than the set target distance. A truly coaxial system should correct this. The landfill data shown in Figure 6 contain the small error due to this effect, but cannot be corrected because not enough auxiliary data were measured at the time. A further complication in making landfill methane measurements is that the methane is generated with a nearly equal amount of CO<sub>2</sub>, so that the density of landfill gas is nearly the same as air. Thus, winds can easily blow landfill gas into an area being measured from another region where it was emitted, because it rises slowly.

The system was taken to Scholl Canyon again on October 8. Three improvements were made in the data-taking strategy. First, the system was tipped at a steeper angle so that it would hit the ground at about 10 m, rather than 14 m. That would make the actual distance to the target less sensitive to the angle between the truck and the ground target, which would serve to reduce the error due to the noncoaxial alignment of the two laser beams. That would also increase the signal strength. The second improvement was that the signal and reference wavelengths would be measured independently. That was done by sending the signal from each laser to a different phase-synchronous detector and further alternating the transmission of each wavelength for a few seconds at a time. This was necessary because there was some crosstalk (~5-10%) between the two channels, even though the beams were chopped 90° out of phase. Third, an attempt could be made to see whether the laser beams were hitting the ground at a greater or smaller distance than the 10 m for which the system was set. Since the concentration-pathlength product is measured, the distance has to be known in order to determine the concentration.

The data from that set of measurements have been reviewed. The general conclusions are that the strong sources of methane were still identified as being in the same locations as during the August 28 measurements, but that the peak concentrations are closer to 50 ppm over 10 m rather than about 125 ppm over 14 m. There are four possible explanations for this difference:

1. The time history of the atmospheric pressure could have been different for the two days. More landfill gas is emitted when the pressure is falling than when it is rising.
2. The temperature of the ground could be lower, causing a slower rate of biogeneration of methane. While data were not obtained, the air during early October was about 5°C colder than late August.
3. Rain could have affected the emission rate. There was some light rain in early October, but it did not penetrate deeply, and had mostly evaporated by October 8.
4. The measurement accuracy may have been better in October than in August, due to better data-taking procedures.

Of these four possible explanations, the first (pressure history) and fourth (measurement accuracy) are the most likely candidates. However, it should be pointed out that measurements of this type may be more valuable in indicating where the places of higher methane emission rates are rather than the actual quantities. The Los Angeles County Sanitation Department made measurements over the same area using a flame ionization detector and found good general agreement of the location of the "hot spots."<sup>(21)</sup>

#### F. Field Measurement of Natural Gas from Underground-Pipeline Leaks

The laser system was taken into Pasadena on the mornings of September 9, 11, and 18 to determine how well it could measure leaks previously located by Southern California Gas Company employees.

The laser system was placed on the sidewalk or street and aimed at the ground behind several leaks. The distance to the target was about 10 to 15 m. The optic axis of the system was about 1.5 m above the ground so the angle was between 5 and 10 degrees from the horizontal.

On September 9, the laser system easily detected two leaks--one from a service connection behind the curb and one from a 1-in.-diameter bar hole in the street--both with gas concentrations on the order of 10% as measured by a flame ionization detector (Century System OVA-88, Foxboro Analytical, Burlington, MA). For the bar hole, the laser system measured 1.7% gas in a 1-in.-diameter plume, from taking the measured concentration-pathlength product and considering it to have a 1-in. diameter. Smaller leaks, previously measured at from 0.02% to 1%, were not detected.

On September 11, the bar hole leak was again measured, as was another leak coming through cracks in the asphalt street and the concrete sidewalk. The leak had been previously measured to be 0.5% with a diameter of 18 ft.

On September 18, the laser system was taken back into Pasadena with a flame ionization detector (FID). The FID measured around 30 ppm hydrocarbons at the bar hole where 10% hydrocarbon had been measured previously. Very little gas was found at any of the other leaks. The weather had cooled a few degrees (5 to 10°F) from September 9 and 11, which may have caused the pipes to contract and seal. More likely, the barhole had allowed a pocket of gas which had accumulated with time in soil of low permeability to vent.

The FID showed that, an inch above the leaks, the gas concentration decreased by an order of magnitude. The wind was a few miles per hour (1-5 mph).

The best signature for these gas leaks appears to be the time variation of the absorption at  $3.3922 \mu\text{m}$  ( $\alpha \text{CH}_4 = 8.8 \text{ atm}^{-1}\text{cm}^{-1}$ ). The fluctuations, due to wind, are on the order of 0.03 to 0.2 sec. Even for cases where the gas concentration-pathlength is high, the fluctuating part of the signal is larger than the steady part of the signal.

#### G. Estimation of System Sensitivity

During the field program in Pasadena, measurements of signal and noise were made that allow the system sensitivity to be estimated. For a time constant of 0.1 sec, and a distance to the asphalt street surface of 13 m, the signal-to-noise ratio was 250 for the signal wavelength. (See Sec. III-D) This translates to a measurement error equivalent to 2.5-ppm methane with a 1-m spatial extent along the line of sight. With easily realizable system improvements (twice as much laser power, slightly higher mirror reflectivities) the signal-to-noise ratio can be increased by a factor of 2, allowing 1.25 ppm-m methane to be detected at 13 m in a fraction of a second.

It should be noted that the global background methane does not, in general, affect the methane leak measurements--as long as the background methane is uniformly mixed and the distance to the target doesn't vary. If the distance to the target varies, then there will be an additional change in the differential absorption of about 0.25% per meter change, or a 1.6-ppm-m change (based on a global background concentration of 1.6 ppm).

As shown in Eqs. 6 and 7, the signal-to-noise ratio depends on several factors. For a given system, the range-dependence and time-dependence are most important. Thus, the sensitivity to methane increases (detection limit decreases) as the range decreases, up to some point a few meters in front of the system. The sensitivity also increases as the integration time increases, but increasing the time constant decreases the rate at which the system can be scanned when searching for leaks.

#### V. DESIGN FOR A PORTABLE LASER REMOTE SENSOR FOR METHANE

The work during Phase I of the contract with GRI on a "Laser System for Global Detection of Natural Gas" has shown that a HeNe laser system has the sensitivity to methane and range required to be useful in the field to search for natural gas leaks from underground distribution pipelines. This was demonstrated with a stationary system pointing at known leaks. Several problems with the present configuration have been noted:

1. Measurement error due to the noncoaxial alignment of the two laser beams,
2. Large weight and size of system, making it difficult to transport and use,
3. Sensitivity to changes in distance to the target,

4. Limited range to target with adequate sensitivity,
5. Small diameter beam at the target,
6. Sensitivity to changes in reflectance of the targets,
7. Amplitude instability of the lasers,
8. Lack of a simple device to indicate where the system is pointing,
9. The data analysis electronics is relatively unsophisticated.

While these problems adversely affect the system performance, they are not permanent impediments to the detection of methane leaks with a HeNe laser system. A consideration of the state-of-the-art in the technology applicable to a HeNe laser system indicates that with some judicious choosing of components and some minor improvements in the state-of-the-art, a portable laser system for the remote detection of methane leaks could be assembled. Likely system components are outlined here.

#### A. HeNe Lasers

One HeNe laser can be designed to generate the two wavelengths required for methane detection. A Fabry-Perot interferometer is used to replace the rear mirror. In general, the Fabry-Perot interferometer will transmit (or reflect) only those wavelengths for which its two mirror spacing satisfies the expression:

$$\lambda = \frac{2d}{m} \quad \text{Eq. 8}$$

where  $\lambda$  is the wavelength  
d is the mirror spacing  
and m is an integer.

If one mirror of the interferometer can be moved, the spacing, d, can be changed so that the other wavelength is favored. The mirror spacing should be such that Eq. 8 is satisfied for one wavelength but off by a half wavelength for the other wavelengths. The smallest separation for which this occurs is given by:

$$\Delta\nu = \frac{1}{4d} \quad \text{Eq. 9}$$

where  $\Delta\nu$  is the separation in wavenumbers ( $\text{cm}^{-1}$ ) between the two wavelengths.

For the two HeNe wavelengths, the separation is approximately  $0.9 \text{ cm}^{-1}$ , giving a minimum separation of about 2.8 mm. The mirror separation would have to change by half a wavelength ( $1.72 \mu\text{m}$ ) to allow the other wavelength to be favored.



This idea could be realized by placing the rear mirror of the Fabry-Perot interferometer on a piezoelectric disc and oscillating it at about 1 kHz. This is similar to what has been done by P. Kebedian.<sup>(22,23)</sup> He limited his cavity length to 7" and included both the wavelength control and plasma tube in that cavity, which limited the laser power to 10  $\mu$ W, too low to be useful for remote measurements. By going to a three-mirror cavity,<sup>(24)</sup> the plasma tube can be made long enough to generate 2 mW without limiting the tuning of the laser. This is shown in Figure 7. Lasing conditions would have to be satisfied for each section of the laser, which means that the tuning may be slightly irregular due to the several hundred MHz mode spacing in the long section.

The laser can be built starting with a plasma tube that can be driven with 20 W of electrical power. An invar-stabilized cavity can be constructed that would reduce amplitude instability due to change in cavity length.

A cell containing a small amount of methane might also be inserted into the cavity in order to equalize the gain at both laser wavelengths so that equal power is generated at each wavelength.

#### B. Collector Mirror

The collector mirror should be large, lightweight, and probably off-axis. The mirror used in the present system has a diameter of 14.5 in. (37 cm), and a usable area of 150 in.<sup>2</sup> (0.095 m<sup>2</sup>). This is adequate for a 2mW of laser power and topographic targets out to 15 m. If a system is to work at ranges as great as 30 m, and since the backscattered signal falls off as the inverse square of the distance, a collector area of up to four times that of the present collector might be desirable. (Alternatively, the time-constant could be increased.) The focal length for use in a portable system should be reduced to about 12 in. (30 cm) in order to tightly focus the collected light on the detector. This reduces the effect of changes in distance to the target and allows the transmitted divergence to be as large as 9 mrad (0.5°). The off-axis requirement arises from the need to look for leaks as close as 3 m from the system. If a detector in a large dewar were at a focal point on an axis in the center of the collector, close-in targets would be obscured. It might be possible to have a small-diameter tip on the dewar so that such obscuration can be minimized. In that case, an on-axis collector could be used. If it were off-axis, a special curve would have to be designed for optimum focusing on a detector for a range of targets from 3 to 30 m. A master surface should then be made and used to generate metal replicas. Since the mirror acts more like a "light bucket" rather than an image, the surface quality should be high enough. Alternatively, a Fresnel lens made of Kel-F could be used (Lectric Lites Co., Fort Worth, TX).

#### C. Detector

A 2-mm-diameter InSb detector, cooled with liquid nitrogen to 77 K, and preceded by a cooled narrow-band filter is seen as the optimal configuration to use. Cooling the detector eliminates much of the blackbody radiation associated with higher temperatures and allows the system to be sensitive to backscattered radiation from topographic targets. The dewar need be filled but once a day with liquid nitrogen. The filter further reduces background radiation incident on the detector.

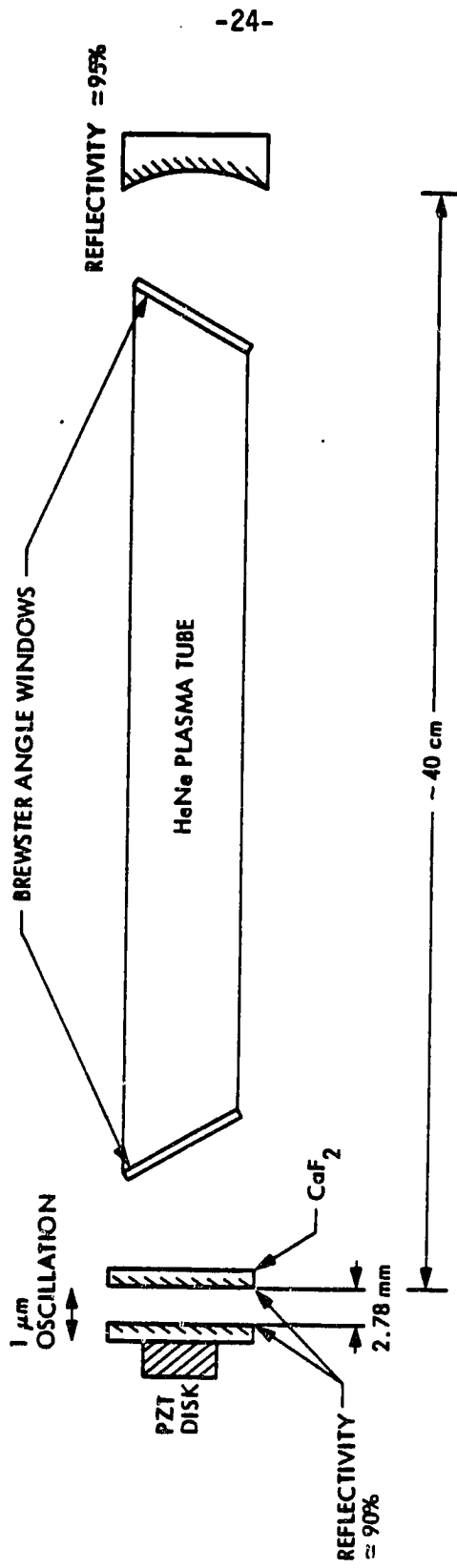


Figure 7. Single HeNe Laser That Generates 3.3911 and 3.3922 μm With High Output Power (~2 mW)

It is possible that cooling could be accomplished with an expanding-gas cryostat (Joule-Thompson effect) although that is not without its problems, too. These include attaching and carrying a large bottle of gas and preventing clogging due to dirt and dust. Another approach is to thermoelectrically cool the detector to 190 K, which would result in a D\* an order of magnitude lower.

#### D. Data System

For small leaks, only one phase-synchronous detector is required using the difference technique (Sec. II C). This can be followed by a device that converts the signal amplitude from the phase-synchronous detector to an audible signal that changes frequency or amplitude with signal strength. The common helium leak detector increases amplitude with increased detection of helium, for example.

#### E. Mounting

The system optical components discussed could be linked with a light-weight yet sturdy frame. Graphite epoxy would be optimal, but is expensive. Alternately, metal frames, such as stainless steel tubing, could be used.

The optical portion of the system would weigh between 10 and 15 pounds. The total system would weigh up to 25 pounds.

#### F. Electrical Power

The system outlined above would draw between 30 and 40 watts of electrical power. This could be supplied from a battery. A lead acid battery that weighs six pounds and supplies 50 W hours of power is available.

### VI. FUTURE SYSTEMS TO MEASURE METHANE AND ETHANE

Systems could be assembled that would have longer range capability and, perhaps, the capability to measure ethane. HeNe lasers can emit up to 20 mW at 3.39  $\mu\text{m}$ . A vehicle system could have a larger collector as well. Assuming a 30-in.- (76-cm) diameter collector, a 20-mW laser, and a falloff in signal as the inverse square of the distance to the target, using Eq. 2, a factor of 4.5x increase in range could be obtained with this system compared with a handheld system with a 14.5-in.- (37-cm) diameter collector and 2-mW HeNe lasers.

If, on the other hand, the technology of tunable lasers improves in the next 2 - 4 years to the point where an inexpensive laser could be used to measure both ethane and methane, and had more power than the HeNe laser, even greater target ranges could be accommodated. Table IV shows the present state-of-the-art of lasers that lase in the 3.1- to 3.9- $\mu\text{m}$  spectral region.

If CW lasers are used and distance to the target should be measured, a GaAs laser range finder could be used. They are available for around \$10,000. (22)

These laser systems are examined in more detail here.

TABLE IV

Lasers in the 3.31- to 3.39- $\mu$ m Spectral Region

| Laser System  | Direct Overlap w/ Strong $C_2H_6$ Line | CW Power | Pulsed Power/Pulse Rate | Size       | Elect. Power | State of Develop.           | Price for First Laser |
|---|--|----------|-------------------------|------------|--------------|-----------------------------|-----------------------|
| 1. Co:MgF <sub>2</sub> + H <sub>2</sub> Raman shift | yes                                    | ---      | ?/30Hz                  | large      | ?            | under dev.                  | \$75K                 |
| 2. CO <sub>2</sub> -tripled                         | yes                                    | ---      | 5mJ/100Hz               | medium     | 300 W        | under dev.                  | \$90K                 |
| 3. CO <sub>2</sub> + CO - summed                    | yes                                    | ---      | ?                       | large      | 500 W        | preliminary research        | \$80K                 |
| 4. Diode  | yes                                    | 0.25mW   | ---                     | medium     | 1 kW         | commercially available      | \$30K                 |
| 5. Frequency difference laser (Nd: YAG + dye)       | yes                                    | ---      | 0.2mJ/10Hz              | large      | ?            | commercially available      | \$75K                 |
| 6. Kr I   | no                                     | low      | ---                     | large      | 4 kW         | ?                           | ?                     |
| 7. N <sub>2</sub>                                   | no                                     | ---      | low                     | large      | ?            | ?                           | ?                     |
| 8. HeNe   | no                                     | 1-20mW   | ---                     | small      | 1-200 W      | commercially available      | \$5-10K               |
| 9. Optical Paramater Oscillator                     | yes                                    | ---      | 1mJ/10Hz                | very large | ?            | university research project | \$200K                |
| 10. HeXe  | no                                     | 0.8mW    | ---                     | medium     | 200 W        | commercially available      | \$8K                  |
| 11. F-center  | yes                                    | 4mW      | ---                     | large      | 20 kW        | commercially available      | \$80K                 |
| 12. Frequency shifted, Nd: YAG-pumped CdSe          | yes                                    | ---      | 20mJ/50Hz               | medium     | 1 kW         | under dev.                  | \$100K                |
| 13. H <sub>2</sub> -cell Raman shifted              | yes                                    | ---      | 2mJ/10Hz                | large      | 2 kW         | commercially available      | \$70K                 |

The Co:MgF<sub>2</sub> plus H<sub>2</sub> Raman-shifted system is being developed at MIT Lincoln Laboratories. <sup>(26)</sup> A Nd:YAG laser is used to pump the Co:MgF<sub>2</sub> laser. The output is in the 1.5- to 2.3- $\mu$ m region. It has generated 150 mJ/pulse at 30 Hz. An H<sub>2</sub> Raman cell is used to shift this radiation to about 3 to 5  $\mu$ m. The crystal is grown only at MIT. It is not likely to be commercially available for several years.

The CO<sub>2</sub> laser can be tripled using two nonlinear crystals. <sup>(27)</sup> Fortunately, the <sup>12</sup>C<sup>16</sup>O<sub>2</sub> ordinary laser lines, <sup>(28)</sup> if tripled, do overlap with the strong ethane lines. <sup>12</sup>C<sup>16</sup>O<sub>2</sub> sequence laser lines would be too weak to be efficiently tripled. <sup>(29)</sup>

Summing CO plus CO<sub>2</sub> would give overlaps with ethane lines. However, the two lasers are not well matched for efficient summing. The pulse width of the CO laser is 50  $\mu$ sec, while that of the CO<sub>2</sub> laser is 0.1  $\mu$ sec.

The diode laser is tunable in the 3.1- to 3.9- $\mu$ m spectral region. The manufacturer offers lasers with power up to 0.25 mW (about a factor of 10 less than that used with the HeNe lasers), which is not quite enough for good signals from topographic targets. A second problem is that diode lasers often lase on several lines simultaneously. This would reduce the power at the ethane line and the absorption from ethane for the laser signal. Also, the diode laser is not an easy laser to use. The calibration of wavelength with electric current often changes when the laser is warmed up from near absolute zero.

The frequency difference laser is commercially available from Quanta Ray. <sup>(30)</sup> In this system, a Nd:YAG laser is doubled, then pumps a dye laser. The Nd:YAG laser radiation and the dye laser radiation are combined in a LiNbO<sub>3</sub> crystal to give the frequency difference, which is tunable in the 3- to 4- $\mu$ m spectral region. One problem with this system is that its frequency is not very stable. Even with intracavity line-narrowing components, the Nd:YAG laser has up to a 0.1-cm<sup>-1</sup> jitter, and the dye laser has about a 0.05-cm<sup>-1</sup> linewidth. The combined linewidth and uncertainty of 0.15 cm<sup>-1</sup> is very close to the line width of the ethane lines (0.2 cm<sup>-1</sup>) so that the laser could not be counted on to hit the center of an ethane line each time. Thus, the effective absorption coefficient of ethane would drop by a factor of 2 or so, making detection less likely. Also, according to the manufacturer, the expected laser output with a Nd:YAG amplifier is only 0.2 mW per pulse at 10 Hz. This is equivalent to a 4-mW HeNe laser chopped with a 50% duty cycle. This amount of power is sufficient with topographic targets and a 14.5-in. diameter receiver out to a range of 25 to 50 m. It is unlikely to scale well to where it could be used in an airplane. Also, the system is still difficult to use.

The neutral krypton laser and the nitrogen laser have a few low-powered lines in the 3.3- to 3.4- $\mu$ m spectral region that don't overlap with the strong ethane lines.

The optical parametric oscillator laser has been under development by Professor Robert Byer at Stanford University for over 10 years. <sup>(31)</sup> It is a Nd:YAG-pumped LiNbO<sub>3</sub> crystal system. It has problems with complexity that may relegate it to the laboratory indefinitely.

The HeXe has one line at  $3.3666 \mu\text{m}$  ( $2969.5 \text{ cm}^{-1}$ ) that is about  $1 \text{ cm}^{-1}$  away from a strong ethane line at  $2970.5 \text{ cm}^{-1}$ . HeXe has been Zeeman-tuned at  $3.508 \mu\text{m}$  by  $7 \text{ cm}^{-1}$  in a field of 70 kG.<sup>(32)</sup> Thus, approximately 10 kG would be required to tune the laser to hit the ethane line. The best that permanent magnets can do is about 3.6 kG. Conventional and superconducting solenoids would be too large for a convenient field system.

The F-center laser is commercially available from Burleigh Instruments.<sup>(33)</sup> A crystal containing color (F) centers is cooled to liquid nitrogen temperature and pumped with a krypton laser. It can generate about 10 mW of tunable radiation from 2.2 to  $3.5 \mu\text{m}$ , with a linewidth of 1.5 MHz or 1.5 GHz ( $1 \text{ cm}^{-1}$  equals 30 GHz). The wavelength can be dithered at rates of 100+Hz so that a "difference" lock-in detection technique can be used for weak ethane signals. This appears to be the best currently available laser system for remote ethane detection. Its primary drawback is that it is a large system, containing a krypton laser that is about 2 m long and draws 3 kW, and an F-center laser that is about 0.7 m long.

The frequency-shifted, Nd:YAG-pumped CdSe laser system is under development at MIT Lincoln Laboratories.<sup>(34)</sup> The CdSe crystal gives tunable radiation in the 1.4- to  $2.4 \mu\text{m}$  region. This can be sent to two  $\text{MgF}_2$  crystals which can be combined in another crystal to give the difference frequency. It is expected that this laser will generate 20 to 50 mJ per pulse in the 3- to  $4 \mu\text{m}$  spectral region at 50 Hz. Although it would have some of the line position uncertainty and linewidth problems of the dye laser difference frequency laser, it should have about 500 times the average power. It is estimated that this laser will be demonstrated in the laboratory in about a year, and will be commercially available in 3 to 5 years.

The  $\text{H}_2$ -cell can be used to Raman shift a Nd:YAG-pumped dye laser.<sup>(35)</sup> Its efficiency drops off as the inverse fourth power of the wavelength, so the efficiency in the infrared range is rather low--perhaps 5% at  $3.3 \mu\text{m}$ . It is expected to be less efficient there than the difference frequency laser.<sup>(36)</sup>

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