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LASER DIAGNOSTIC TECHNIQUES AT POLY-  
TECHNIC INSTITUTE OF NEW YORK AERO-  
SPACE FACILITIES

S. Lederman, et al

Polytechnic Institute of New York

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POLYTECHNIC INSTITUTE OF NEW YORK  
Department  
of  
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May 1974

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## TABLE OF CONTENTS

| <u>Section</u> |                                                       | <u>Page</u> |
|----------------|-------------------------------------------------------|-------------|
| I              | Introduction                                          | 1           |
| II             | The Measurement of Density and Concentration          | 3           |
| III            | Laser Raman and Doppler Measurements                  | 5           |
| IV             | Experimental Facilities and Some Experimental Results | 9           |
| V              | Discussion of Results                                 |             |
| VI             | References                                            | 14          |

## LIST OF ILLUSTRATIONS

| <u>Figure</u> |                                                                                                                                   | <u>Page</u> |
|---------------|-----------------------------------------------------------------------------------------------------------------------------------|-------------|
| 1             | Schematic of Experimental Apparatus for Raman Cross-Section Measurement                                                           | 16          |
| 2             | Measured Raman Stokes Intensity of Carbon Dioxide ( $\text{CO}_2$ ) as a Function of Pressure Measured in Air at Room Temperature | 17          |
| 3             | Measured Raman Stokes Intensity of NO in the Presence of Water Saturated Nitrogen vs. NO Partial Pressure (NO-Nitric Oxide)       | 18          |
| 4             | Schematic Diagram of Experimental Apparatus (Co-Axial Jet)                                                                        | 19          |
| 5             | Data Acquisition and Processing                                                                                                   | 20          |
| 6             | Instantaneous Specie Concentration of $\text{CO}_2$ in a Turbulent Jet at $x/D=5$                                                 | 21          |
| 7             | $\text{CO}_2$ Concentration - $x/D=20$                                                                                            | 22          |
| 8             | Temperature Profile in a Methane $\text{CO}_2$ Flame                                                                              | 23          |
| 9             | Apparatus for Resonant Raman Scattering Experiments                                                                               | 24          |

## LIST OF SYMBOLS

|            |                                                       |
|------------|-------------------------------------------------------|
| $A$        | complex electric field amplitude of the incident wave |
| $a$        | particle diameter                                     |
| $C_1$      | constant                                              |
| $c$        | velocity of light                                     |
| $D$        | beam diameter                                         |
| $d$        | diameter of Airy disc                                 |
| $E$        | transition probability                                |
| $f$        | focal length                                          |
| $f_d$      | Doppler frequency                                     |
| $f_o$      | incident frequency                                    |
| $k$        | Boltzmann's constant                                  |
| $k_o, k_s$ | unit vectors in the incident and scattered directions |
| $M_1$      | induced dipole moment                                 |
| $N$        | number of scatterers                                  |
| $T$        | temperature                                           |
| $\alpha'$  | isotropic part of the change of the polarizability    |
| $\nu'$     | anisotropic part of the change of the polarizability  |
| $\lambda$  | wavelength                                            |
| $\nu$      | wave number                                           |
| $\chi$     | fraction of the anisotropic scattering                |

### Subscripts

|           |               |
|-----------|---------------|
| $A$       | Anti-Stokes   |
| $k, n, r$ | energy levels |
| $S$       | Stokes        |
| $o$       | incident      |

## I. INTRODUCTION

With the modern development in aeronautical sciences a major effort has been directed at the problem of obtaining meaningful measurements in the ground-based experimental facilities. As is well-known, some of the problems of diagnostics of flow fields have been studied since the inception of the science of fluid dynamics. As a result of these efforts, some very sophisticated techniques have been developed. Some of these techniques became standard in the field. The optical techniques such as schlieren, shadow, interferometry and holography are capable of providing excellent qualitative insights into some flow fields. While there are a number of methods which permit the extraction of quantitative density data from the above techniques, they are generally extremely complicated, time-consuming, expensive, and prone to too many errors to be of any practical importance. The general applicability of the above techniques is limited by the shortcomings of all diagnostic techniques which depend on the index of refraction.

Other techniques such as the electron beam fluorescence technique, emission and absorption techniques, while capable of providing quantitative data, have limitations on their range of applicability or resolution.

Probe techniques, such as pressure temperature, heat transfer, hot wire anemometer, chromatography, etc., can provide excellent quantitative information on the parameters of a given flow field. However, the mere presence of a probe may alter the given flow field and distort the information one is seeking. Moreover, in some



cases, the ambient conditions in the flow may preclude the use of probes. Extremely high temperatures, pressures, and particulates present in a flow may be highly destructive for any probe.

In view of the above, a search for a measuring technique which could overcome most of the limitations and shortcomings of the standard diagnostics was undertaken.

As is well-known, an ideal measuring technique would permit the attainment of most information, remotely, without disturbing probes, instantaneously and simultaneously.

This we believe can be achieved by the use of the Raman effect. It has been shown<sup>1</sup> that it is possible not only to obtain individual specie concentration measurements in gas mixtures, but also to obtain measurements of specie concentration<sup>2</sup> under adverse ambient conditions which would make other measurement impossible. Furthermore, it was shown<sup>3</sup> that it is possible to apply this method to the remote measurement of pollutants in the atmosphere. This diagnostic technique proved itself to be very effective in the measurements of concentration in an isothermal, low speed turbulent jet and in a methane CO<sub>2</sub> flame<sup>4</sup>.

After a short discussion of some deficiencies of some standard diagnostic techniques, the advantages and limitations of the Raman diagnostic technique is reviewed. Some laboratory arrangements for a number of experimental investigations relating to the applicability of the Raman effect for fluid dynamic measurements are described. Some experimental results obtained in our laboratory are discussed.

## II. THE MEASUREMENT OF DENSITY AND CONCENTRATION

The shadowgraph and schlieren give indications of the second derivative and first derivative of the density in a flow field, respectively. Their main use is, therefore, flow field visualization rather than quantitative density measurements. The interferometer, on the other hand, is a most precise instrument not only for flow visualization but also for direct indication of density variation in a flow field. There is, however, one major drawback when quantitative data are desired. While the interferometer gives a direct quantitative indication of the density changes in a flow field, these changes represent integrated values over the entire thickness of the flow field. This drawback, common also to shadowgraphs as well as to schlieren, makes the above optical methods for quantitative flow diagnostics of questionable value. Even with the most advanced microdensitometric and data processing equipment available, the best one can expect is a two-dimensional map of integrated density points.

For hypersonic tunnels where test section pressures are less than about 1 mm Hg, a density probe has been developed which uses the fluorescence produced by a narrow, well-collimated, high energy electron beam. By focusing the luminescence, which is largely confined to the electron beam region itself, onto an optical slit which is at a right angle to the beam, the light from a small gas volume is selected and can subsequently be measured through the use of a wave selector and photomultiplier. The wave selector can be a spectrograph or a narrow bandpass filter. The intensity of the light is related to the density and can therefore be used as

a direct measure of the gas density at a given point in space and in time. This kind of probe is very useful as long as the gas density is low. As the gas density increases and the time between collisions becomes comparable to the lifetimes of the excited states of the molecules, a measurable fraction of the electron beam excited molecules collide with other molecules, giving up part of their energy in a form other than radiation. This effect, known as collisional quenching, which is temperature as well as density dependent, limits the range of applicability of the electron beam density probe. It was found that for the nitrogen line  $3914\text{\AA}$  generally used in hypersonic flows, there is no quenching effect up to a test section static pressure of  $100\mu\text{ Hg}$ . The applicability of the electron beam probe for density measurement has thus an upper limit of about  $100\mu\text{ Hg}$ . As far as the lower limit is concerned, this would be set by the signal-to-noise ratio of the photomultipliers and associated electronic processing equipment.

The question of specie concentration measurement is generally dealt with through the application of gas chromatography. This type of measurement, which is based on the thermal conductivity of individual gases, is generally limited to a mixture of two gases. Furthermore, due to the underlying principles of this diagnostic technique, a single point measurement requires a relatively long time to perform. This technique is therefore limited to two specie mixtures in a steady state equilibrium. High frequency time varying concentration measurement cannot be obtained. A very important parameter generally required in fluid dynamic measurements is the velocity. In this respect the hot wire anemometer

used for the first time about 50 years ago was developed into a very sensitive and sophisticated instrument. Again, the underlying principle of operation of the hot wire is heat transfer. This instrument is capable of providing measurements from a few feet/sec. to supersonic velocities and for laminar-to-turbulence frequencies of several hundred thousand cycles. There are, however, limitations and shortcomings associated with this instrument. Since the sensing element is basically a heat transfer element, it is sensitive to temperature, density, viscosity, specific heat, conductivity, geometry, etc. This in itself makes a measurement very tedious and complicated. If one considers further the problems associated with compensating for the presence of the probe and the disturbing effects any probe may have on the flow, it is seen that it would be desirable to have a technique which would be totally or nearly independent of all these effects. In what follows, it is believed, a partial answer to the problems of density, concentration, and velocity measurements can be found.

### III. LASER RAMAN AND DOPPLER MEASUREMENTS

As indicated previously, the Raman scattering technique is capable not only of supplementing the above techniques, but also of complimenting them. The theoretical background on which the Raman diagnostic technique is based has been discussed in a number of books and scientific articles<sup>5-11</sup>. The application of the Raman effect towards the diagnostics of flow fields is based upon the premise that most molecular species present in the flow are Raman active, and have their particular characteristic frequency shifts. At this point it is worthwhile to note that all homonuclear diatomic

molecules like  $O_2$ ,  $N_2$  or  $H_2$  are infrared inactive while they are all Raman active. This is one of the very important properties of the Raman effect which makes its application to aerodynamic flow field diagnostics very useful.

The relation between the scattered radiation and the incident radiation is

$$I_{s,A} = C_1 N (\nu_0 \pm \nu)^4 |E_{kn}|^2$$

where

$$E_{kn} = \frac{1}{h} \sum_{\nu} \left\{ \frac{(\bar{A}\bar{M}_{kr}) \bar{M}_{rn}}{\nu_{rk} - \nu_0} + \frac{\bar{M}_{kr} (\bar{A}\bar{M}_{rn})}{\nu_{rn} + \nu_0} \right\} \quad (1)$$

In terms of the Placzek polarizability theory the scattered intensity is given by

$$\frac{I_{s,A}}{I_0} = C_2 \frac{N (\nu_0 \pm \nu)^4 (\alpha'^2 + \frac{7}{45} \chi_{\nu}^2)}{\nu (1 - \exp[-\frac{h\nu}{kT}])} \quad (2)$$

The ratio of the Stokes to Anti-Stokes Raman intensity is given by

$$\frac{I_s}{I_A} = \left( \frac{\nu_0 - \nu}{\nu_0 + \nu} \right)^4 \exp \frac{h\nu}{kT} \quad (3)$$

The above equation presents the essential features of the Raman effect. With the additional feature of being extremely fast (of the order of  $10^{-14}$  sec. for visible light illumination), it is obviously a very useful diagnostic tool for a wide variety of physical and chemical phenomena. It permits one to obtain quantitative information on the individual specie concentration, on the state of ionization, on the temperature of a given specie in a mixture at a

point in a flow field without probes and their disturbing effects. The measurements can be obtained remotely, instantaneously, and simultaneously. The number of points measured simultaneously is only limited by the number of monitoring stations. These qualities place this diagnostic technique on the list of ideal methods. In reality, there are a number of difficulties encountered. One of the major difficulties is the extremely low equivalent Raman scattering cross-section. While this can partially be overcome by high laser power and high gain photomultiplier tubes, there are limitations on the minimum concentration that can be resolved. Discussions concerning these and other features of laser Raman diagnostics are given in Refs. 1, 2 and 4.

The combination of the laser and the Doppler law resulted in the development of a new type of anemometer, the so-called laser Doppler velocitimeter, LDV for short. The LDV relates the velocity of a solid particle in a fluid (seeded or naturally present) to the frequency shift of the scattered light from the primary frequency. This frequency shift is given by

$$f_d = \frac{f_o}{c} \bar{V}(\bar{k}_s - \bar{k}_o) \quad (4)$$

Although the scattering phenomenon responsible for the LDV operation is essentially given by the Mie formula (which depends upon  $\frac{\alpha}{\lambda} \gg 1$ ,  $\alpha$  being the particle diameter), the frequency shift does not depend on the size of the particulate. The LDV has been treated in great detail in many publications too numerous to list here.

The LDV is essentially more a problem in data processing than in the basic operation. The velocity as measured by an LDV is essentially the velocity of a particle in a scattering volume. The scattering volume is therefore an important consideration in designing an LDV. This value is determined by the optics of the transmitter, receiver, and the size of the laser beam, and is limited by the Airy disc given by

$$d = 1.22 \frac{\lambda f}{D} \quad (5)$$

It is therefore obvious that scattering values much smaller than hot wire anemometer probes are usually possible.

In general, compared to the hot wire anemometer, the LDV is more accurate to a wide range of velocities, is linear, independent of other velocity components in the flow, is an absolute measurement depending on the geometry, optics, and laser used. It thus does not need calibrations. It has better spatial resolution, and what is most important, it does not perturb the flow.

A basic assumption underlying the application of the LDV is that the scattering particles follow faithfully the velocity of the fluid under investigation. This, however, is not always the case. In turbulent flows at high velocity and high frequency turbulence, the particle inertia effect alone may be considerable. The velocity ratio of a particle and the flow would fall by about 10% for a particle of  $1\mu$  in diameter at a frequency of 20 kc or a particle of  $3\mu$  diameter at 2.7 kc or  $10\mu$  at 250c/sec. This points out some difficulties of an LDV. Another difficulty is the broadening of the Doppler line due to the finite residence time of the

scattering particle and the random distribution of scatterers.

#### IV. EXPERIMENTAL FACILITIES AND SOME EXPERIMENTAL RESULTS

The experimental efforts concerning laser diagnostics at PINY are manifold. They range from the experimental determination of the equivalent Raman scattering cross-section of relevant molecular species to the determination of concentration of mixtures of gases at best, the determination of concentration of particular species in jets, the measurement of temperature in flames, the determination of velocity fluctuation in a jet using an LDV, the determination of particulates in a flow, the application to pollution monitoring, and investigation of the resonant Raman effect. The basic experimental apparatus for the equivalent Raman cross-section measurement is shown in Fig. 1. This apparatus is also used for the determination of particular species in mixtures<sup>1</sup>. The procedure for the determination of the equivalent Raman scattering cross-section is very simple. The chamber is filled with a gas whose cross-section is known; the pressure and temperature are noted and the scattered intensity measured. The same chamber is filled with a gas of unknown cross-section and under the same conditions of pressure and temperature. The scattered intensity is again measured. From the two measurements the unknown equivalent scattering cross-section is obtained. The scattering cross-section for some species thus determined is for  $\text{CO}_2$ ,  $\sigma=2$  and  $\text{NO}$ ,  $\sigma=0.55$  referred to  $\text{N}_2$ .

Figure 2 presents the Raman intensity measurements as a



function of pressure in the chamber of a single specie and Fig. 3 presents the scattered intensity as a function of NO concentration in water saturated nitrogen atmosphere. It should be noted here that the measurement of NO in the presence of water is extremely difficult using any other method. Figure 4 shows a schematic diagram of the experimental apparatus used to obtain simultaneously the concentration, temperature, and velocity of an axisymmetric turbulent mixing jet and a methane  $\text{CO}_2$  flame. This apparatus consists of a Ruby laser of 150 mW peak power capable of single pulse as well as repetitive operation at a pulse repetition rate of 6 ppm, a 50 mW Jodon He-Ne laser for the LDV, spectrographs including a Spex 1406 double-monochromator, data acquisition and processing equipment including an on-the-line PDP8 computer and all the necessary control equipment for the maintenance of the given condition of the jet flow and flame. In view of the relatively high scattering signals obtained under the experimental conditions described here, the data acquisition equipment used here is quite simple. The signal from the photomultiplier tube is amplified and fed into a linear gate and hold module, where the signal is integrated and stretched such that it can be fed into a multiplex analog to a digital converter which is compatible with the PDP8 digital computer. The linear gate is opened only for a predetermined time, such that only the scattered signal is integrated. The errors which may be caused by noise signals in the photomultiplier or amplifier can thus be minimized. The data acquisition system is capable of accepting 8 signals. One of

those signals is the laser signal used to trigger the gate generator, initiate the data processing cycle, and provide a normalization signal for the received signals. A schematic diagram of the data acquisition and processing system is shown in Fig. 5. Using this system specie concentration profiles on a turbulent mixing jet as well as temperature profiles of methane  $\text{CO}_2$  flame were obtained. Some of the results are shown in Figs. 6, 7, and 8.

The LDV used in a laboratory is of the dual scatter single velocity component type. The signal is displayed on CRT frequency analyzer. At the present time, work is in progress on the assembly of a Doppler data processing system which would permit better data acquisition and the construction of a proper particle seeding system for the jet and flame.

The features of the Raman scattering diagnostic technique, as indicated above, suggests the use of this technique for remote air pollution monitoring and measuring. As shown in Ref. 3, the major drawback of this technique is the extremely small scattering cross-section and the attenuation of the signals at large distances. While short distance (below 2 km) remote, single-ended measurements of pollutants of high concentration can be easily made using the normal Raman effect, and single pulse operation large distance (over 2 km) measurements of very low pollutant concentration measurements may require not only repetitive operation of the laser with statistical data processing but also further developments in laser technology, including the application of the resonant Raman effect. To that end a research effort has been

initiated at PINY. As evident from Eq. (1), when the incident frequency  $\nu_0$  approaches the electronic absorption frequency  $\nu_{rk}$  of the particular molecule, a resonance occurs. Under certain conditions the scattered intensity may increase by several orders of magnitude. This would of course be very desirable. One could either resolve a lower concentration or resolve the same concentration at much greater distances. There are, however, difficulties encountered. The laser must be continuously tunable in order to find the regions of strong resonance enhancement of the species of interest. Once those regions are known, the incident wavelength must be carefully controlled to maintain a precise separation from resonance.

At resonance, the absorption of the incident light results in fluorescence emission and not Raman scattering. While fluorescence emission is generally very intense and characteristic of a given molecule, collisional processes render the fluorescence emission inapplicable for quantitative concentration measurements.

Figure 9 represents an experimental apparatus used for the investigation of the resonant Raman effect. It is constructed to minimize the path of the incident and scattered radiation through the gas under investigation, thus decreasing the attenuation of the radiation. The laser is a nitrogen laser. The gases being investigated are the halogen gases.

## V. DISCUSSION OF RESULTS

As is evident from the data in Fig. 2, the Raman scattering technique is capable of determining the concentration of a specie over a relatively wide dynamic range. The response is linear with the concentration and the accuracy is in most cases adequate. Furthermore, the data of Fig. 3 represents a measurement which is extremely difficult to obtain by any other means. The response is again linear independent of the presence of other species in the mixture.

Figure 6 shows the concentration measurements of  $\text{CO}_2$  in an axisymmetric turbulent jet superimposed on the concentration data obtained using a conductivity cell. At a first glance it appears that the scatter of the data is excessive. In reality, this scatter is an indication of the turbulent character of the flow. This is confirmed by the data of Fig. 7, where statistical averages 50 pulses per point are plotted. These data approach the data as obtained using conductivity cells. It must be noted that the conductivity cell used in this experiment was only capable of measuring mean values of concentration.

Figure 8 shows the mean temperature of a methane flame as obtained using the ratio of the Stokes to the Anti-Stokes vibrational line. Superimposed is the temperature as obtained by a thermocouple.

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Interscience Publishers, New York, 1959.

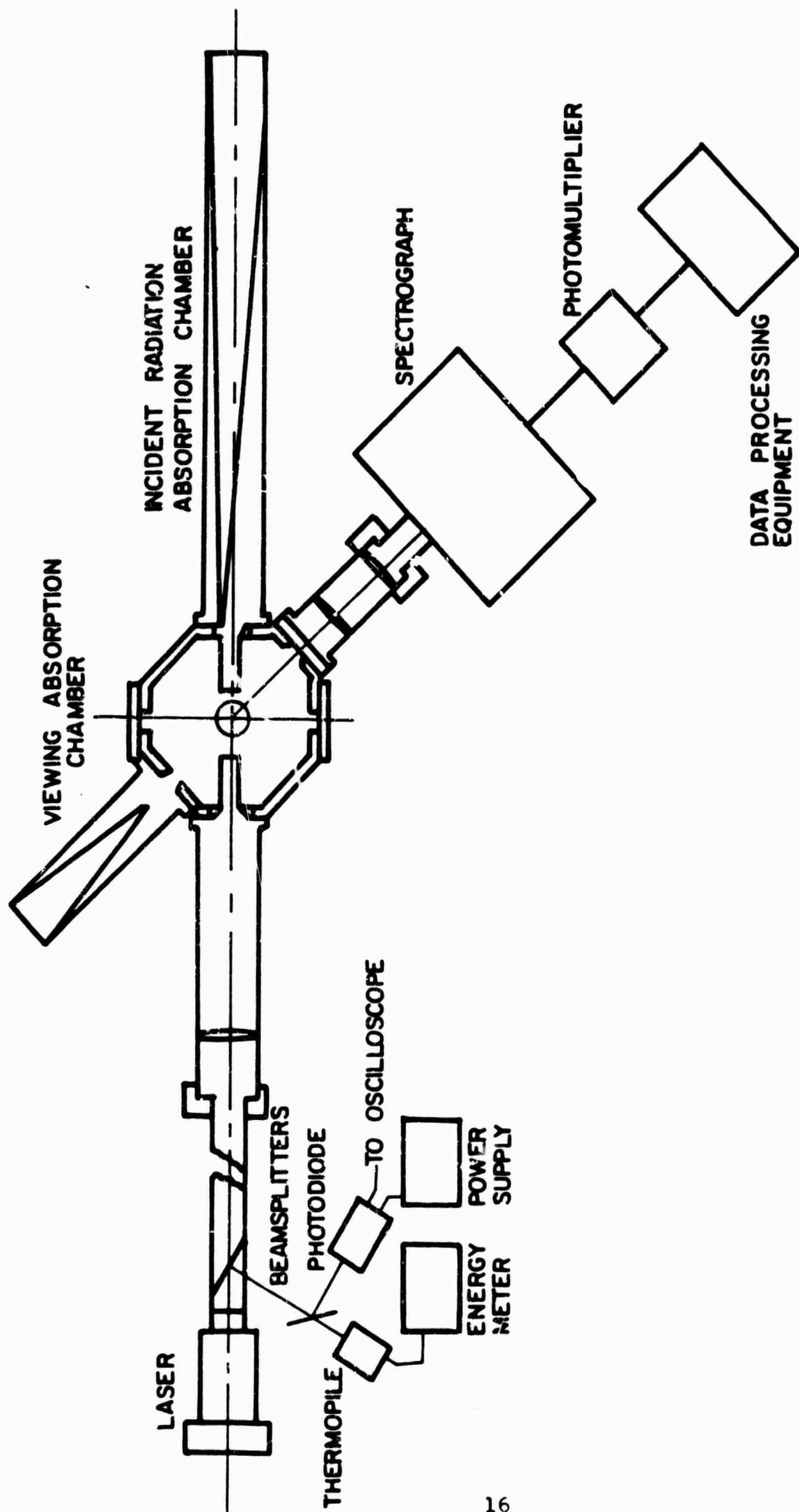
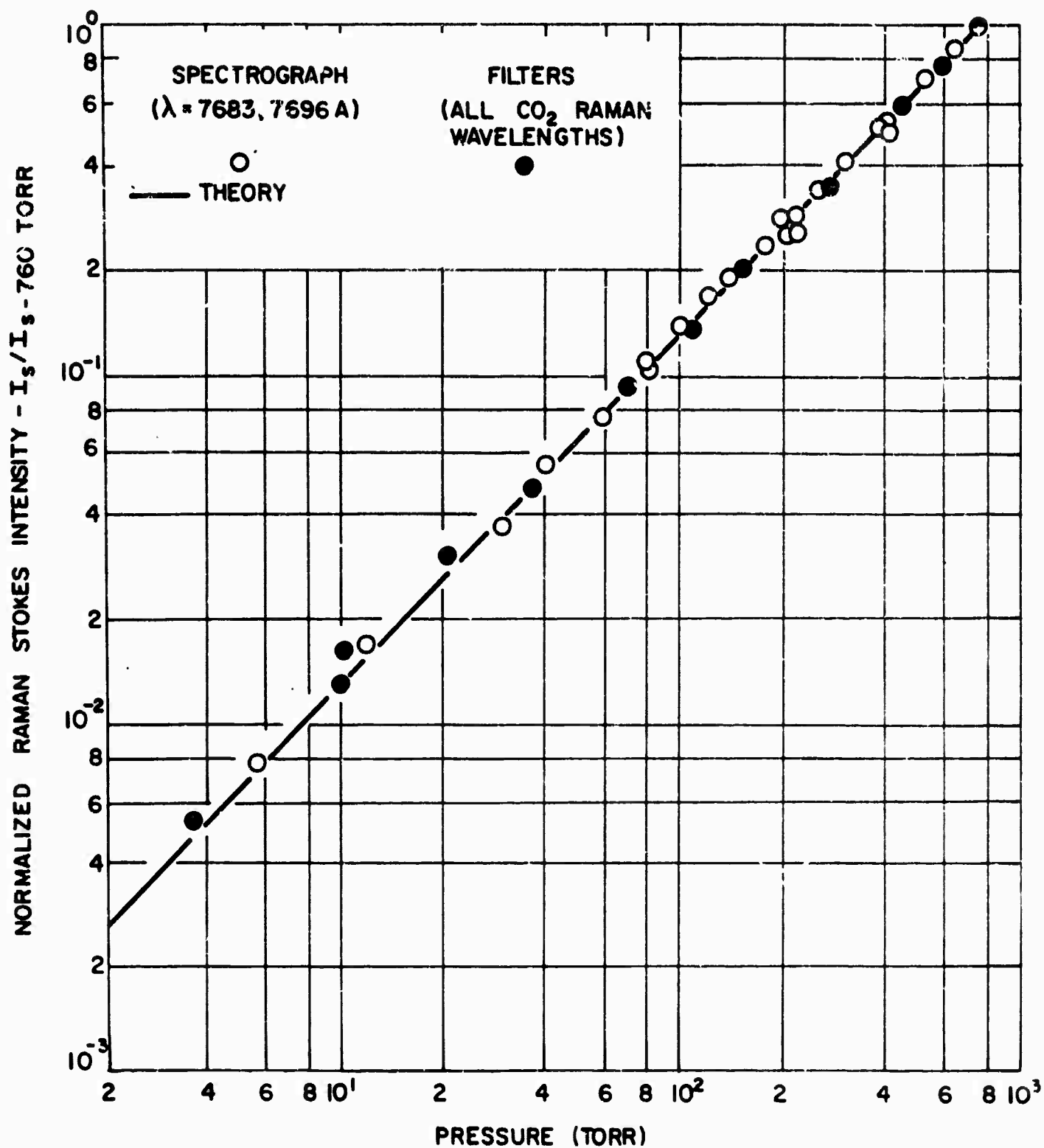


FIG. 1 SCHEMATIC OF EXPERIMENTAL APPARATUS FOR RAMAN CROSS-SECTION MEASUREMENT



**FIG.2 MEASURED RAMAN STOKES INTENSITY OF CARBON DIOXIDE ( $\text{CO}_2$ ) AS A FUNCTION OF PRESSURE. MEASURED IN AIR AT ROOM TEMPERATURE**



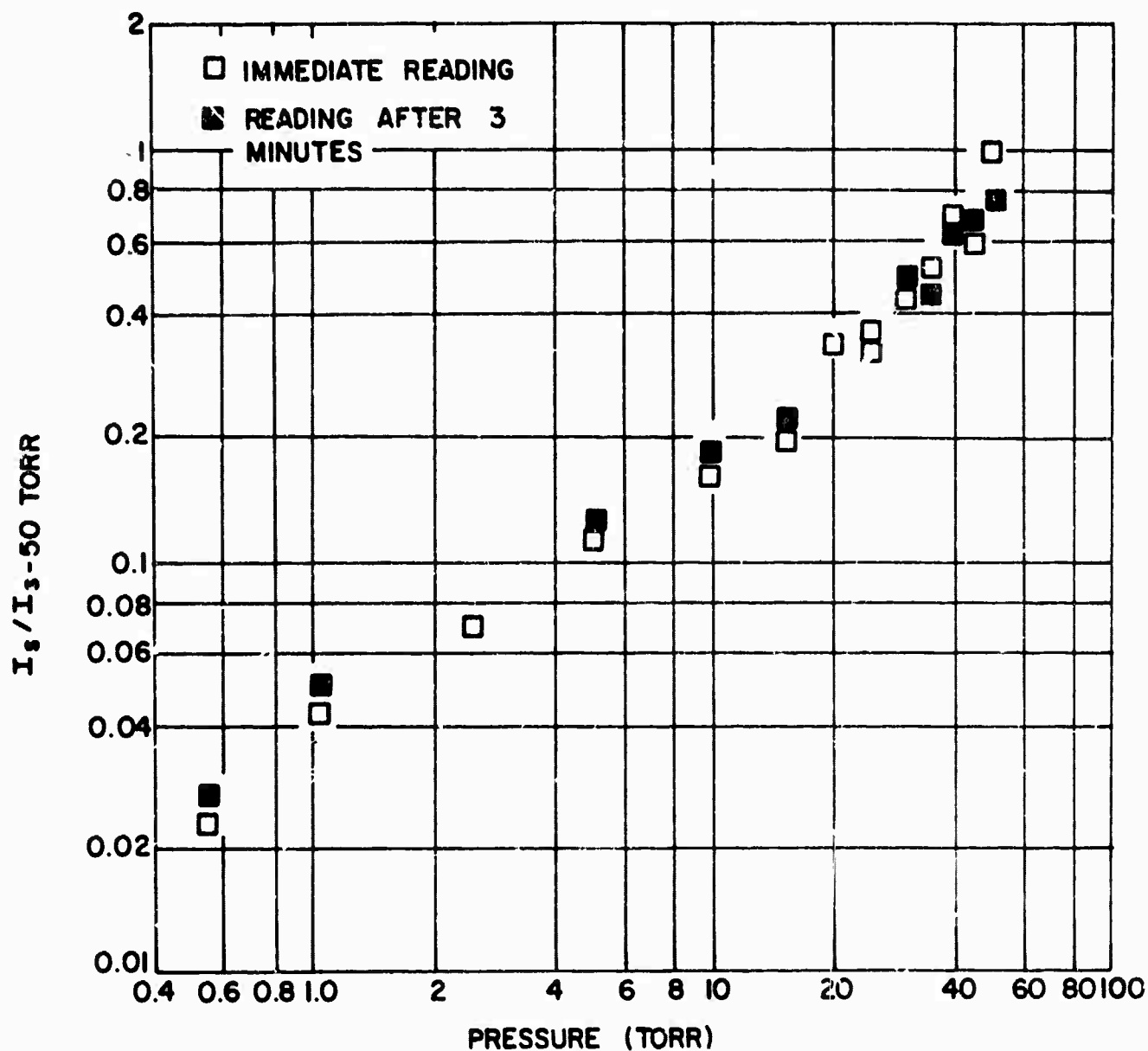


FIG. 3 MEASURED RAMAN STOKES INTENSITY OF NO IN THE PRESENCE OF WATER. SATURATED NITROGEN VS. NO PARTIAL PRESSURE (NO - NITRIC OXIDE)

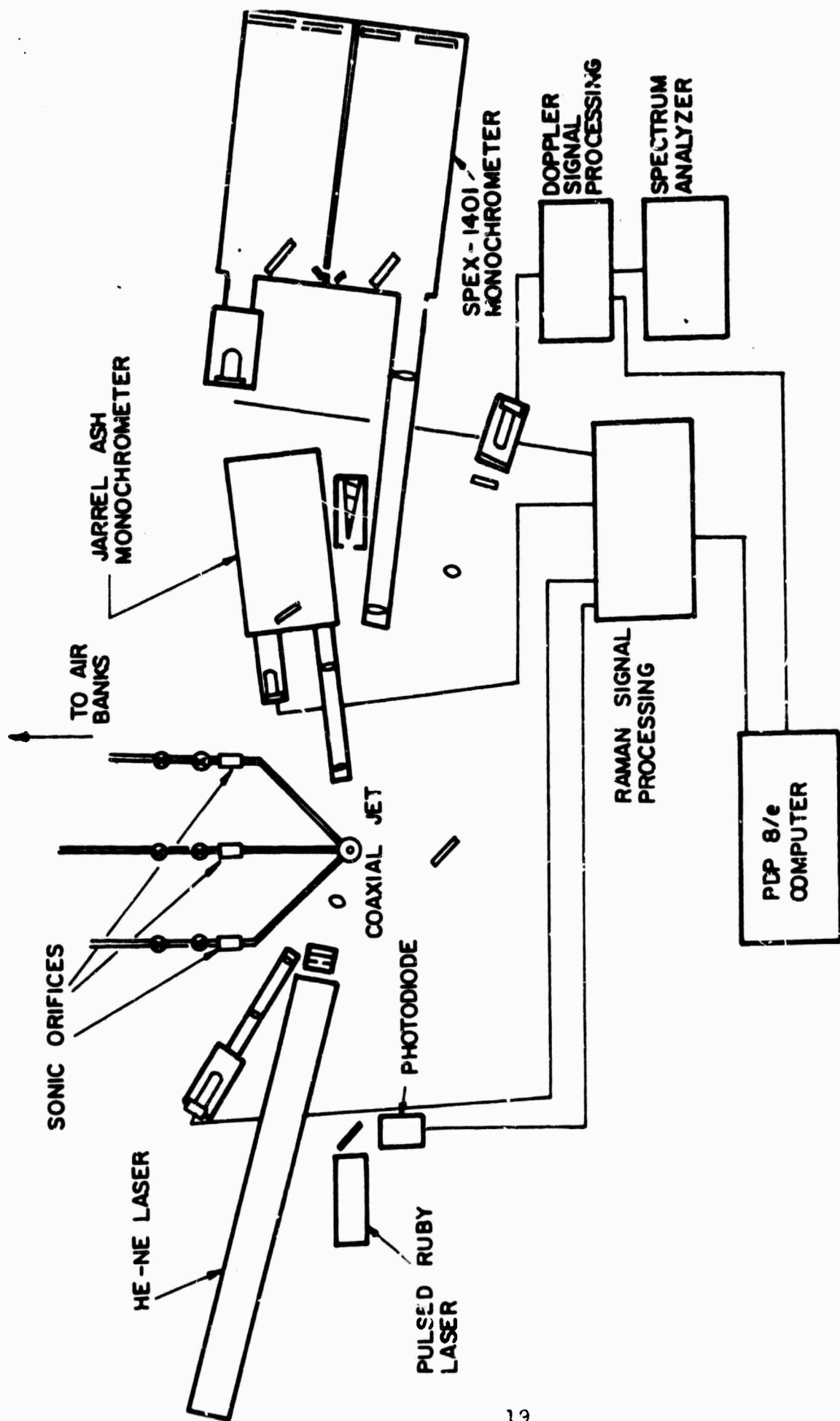
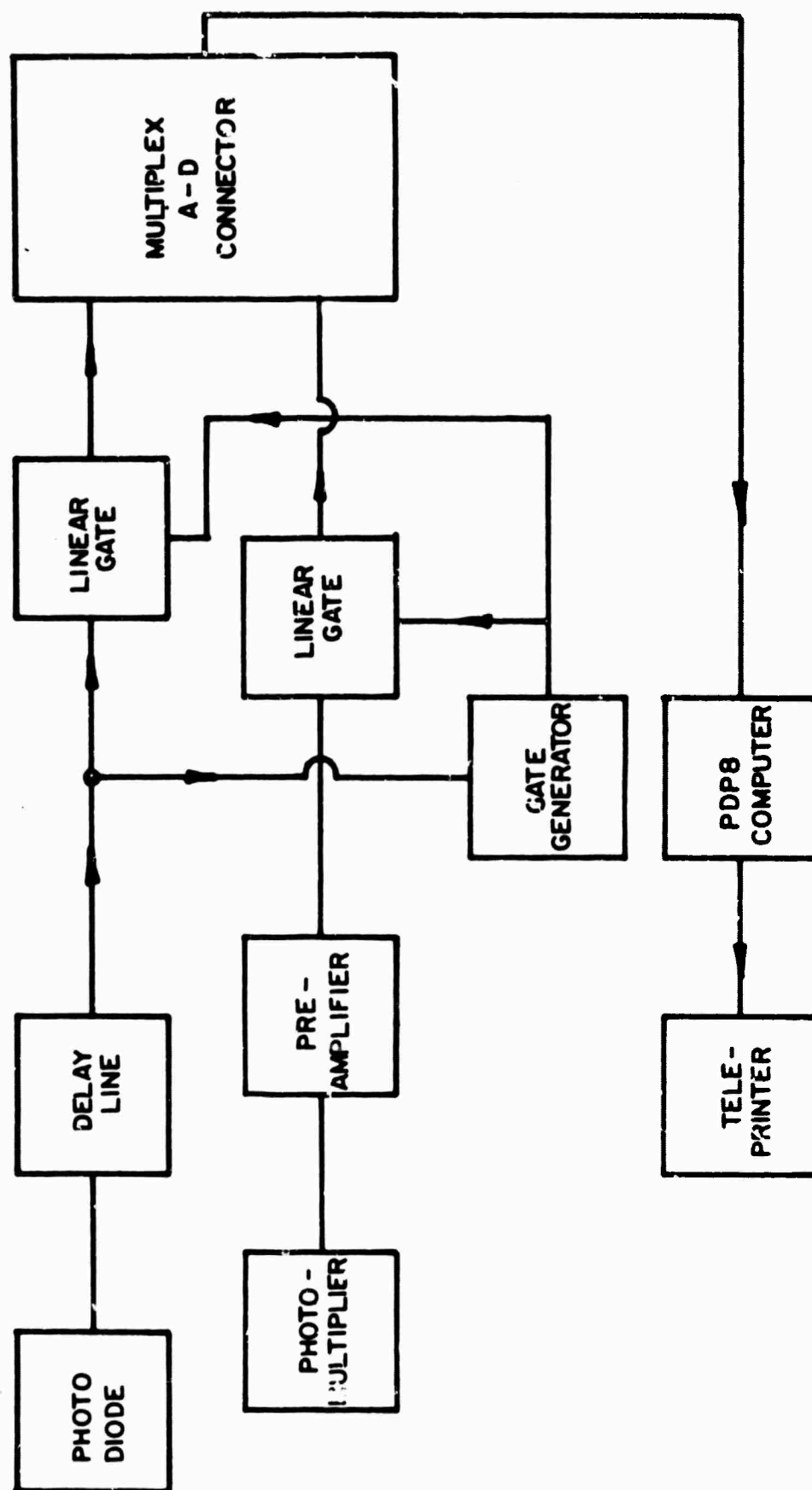


FIG. 4 SCHEMATIC DIAGRAM OF EXPERIMENTAL APPARATUS  
(CO-AXIAL JET)



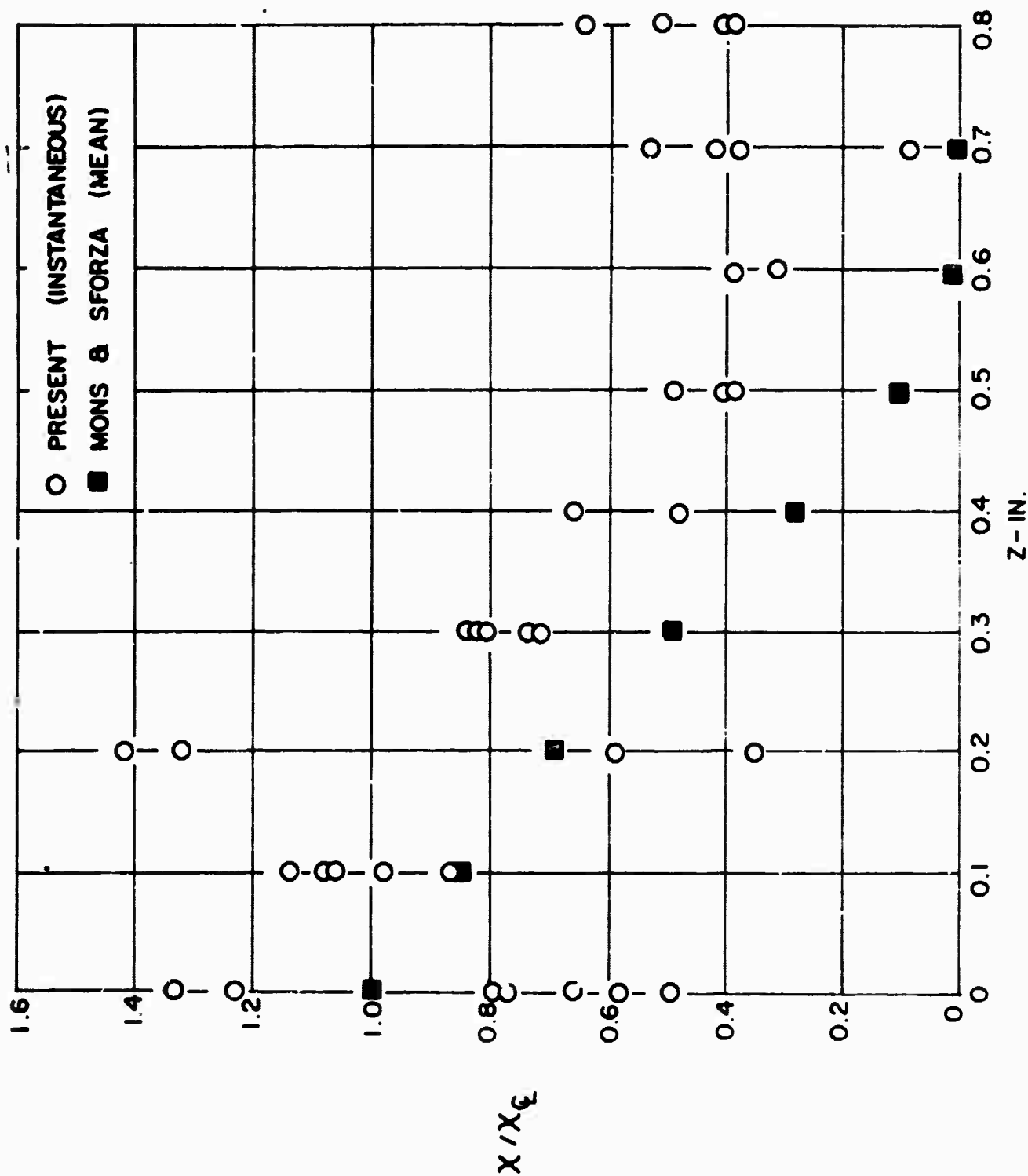


FIG. 6 INSTANTANEOUS SPECIE CONCENTRATION OF CO<sub>2</sub> IN A TURBULENT JET AT  $x/D = 5$

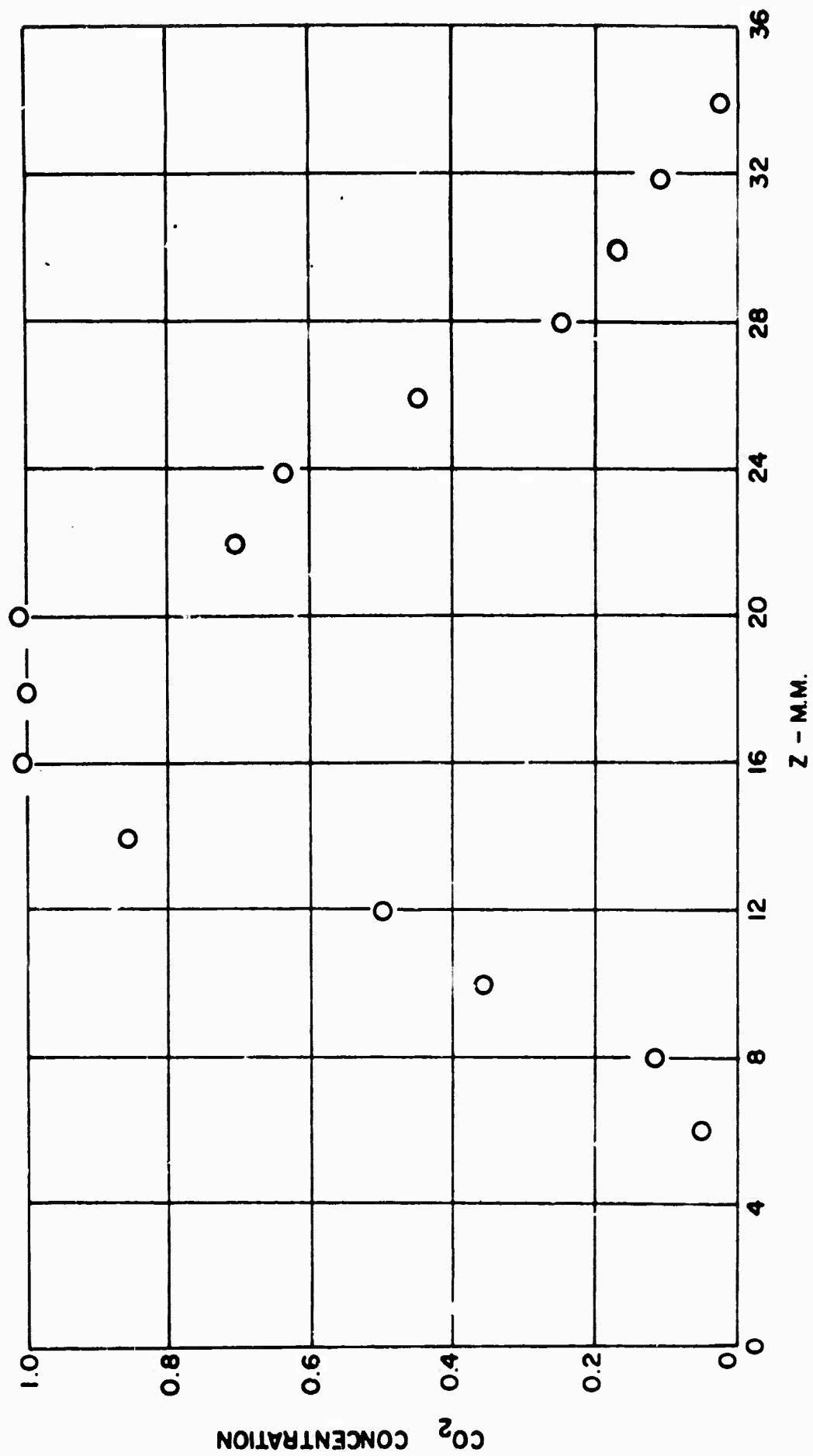
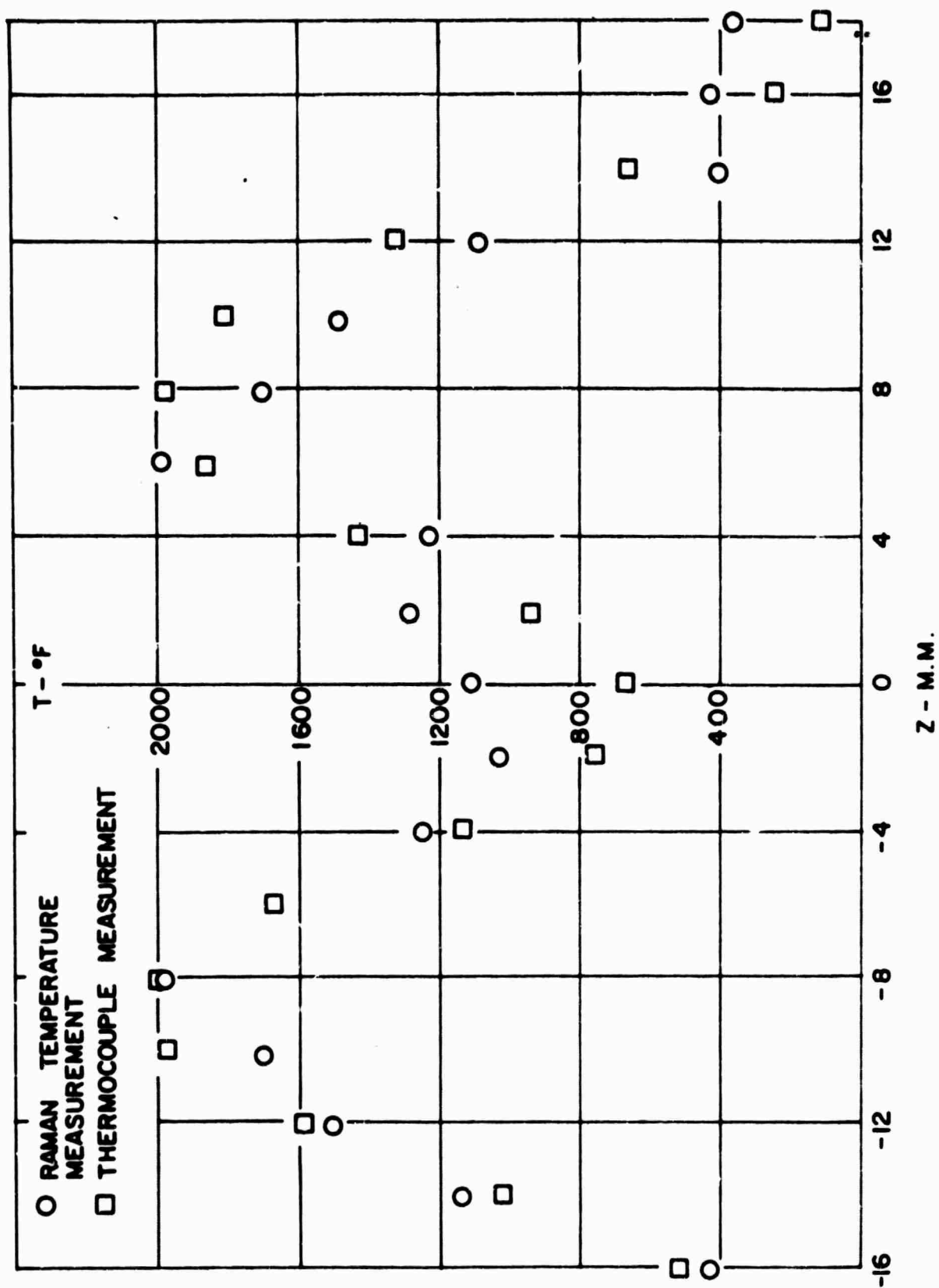
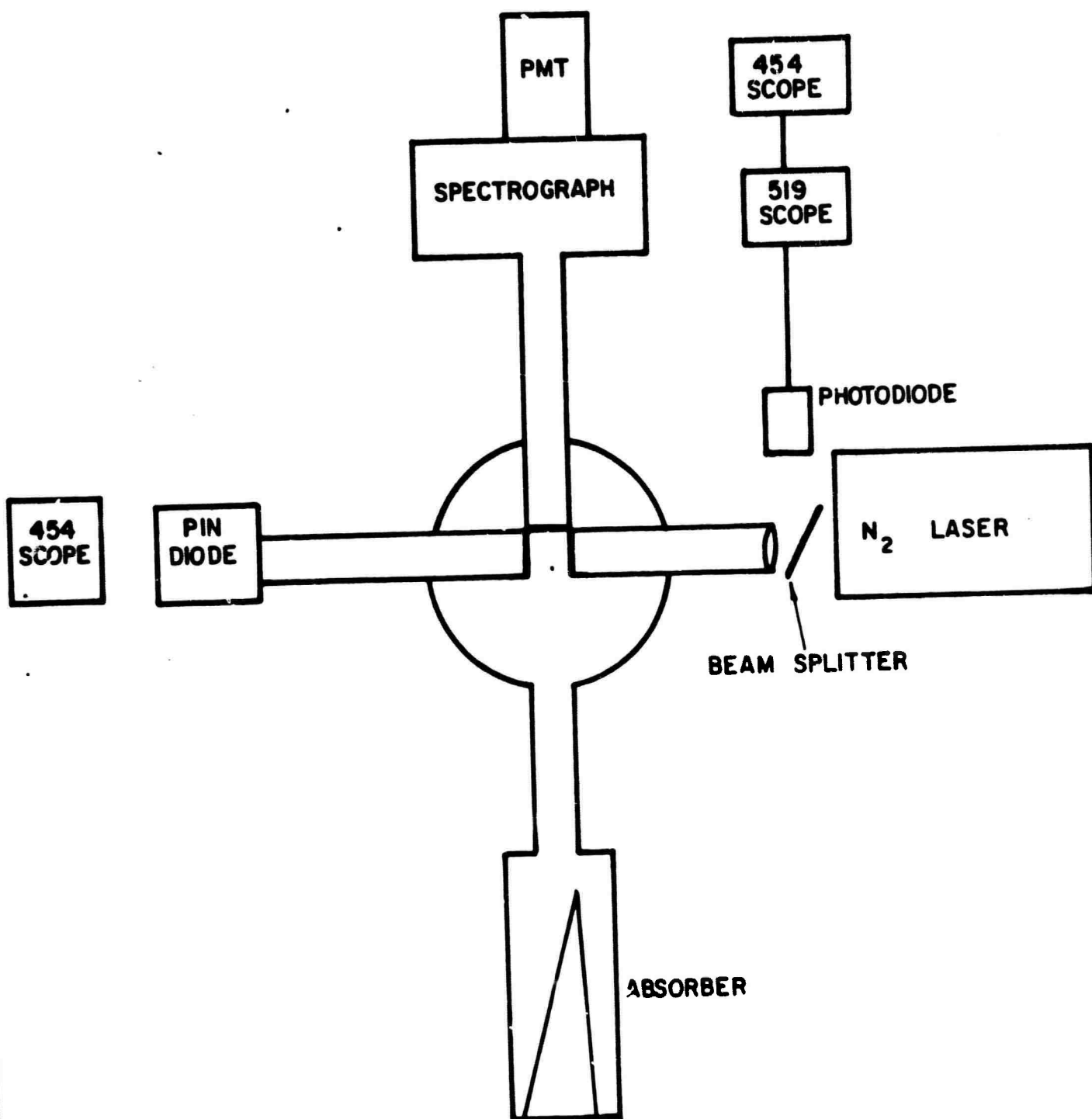


FIG. 7 CO<sub>2</sub> CONCENTRATION -  $x / D = 20$



**FIG.8 TEMPERATURE PROFILE IN A METHANE CO<sub>2</sub> FLAME**



**FIG. 9 APPARATUS FOR RESONANT RAMAN SCATTERING EXPERIMENTS**