Int. J. Heat Mass Transfer. Vol. 17, pp. 1479–1486. Pergamon Press 1974. Printed in Great Britain

TEMPERATURE AND SPECIE CONCENTRATION MEASUREMENTS IN A FLOW FIELD

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(*Received* 30 *April* 1973 *and in revised form* 15 *January* 1974)

Abstract—The Raman scattering technique and its application to the diagnostics of flow fields is discussed. It is shown experimentally that the technique is capable of providing not only specie concentration and their temperature in a quiescent mixture of polyatomic gases, but also in a flow field. It is further shown that due to the extremely fast Raman transition, utilizing the simple high laser power pulse technique, the existence of transient fluctuations in the flow field both in terms of specie concentration and temperature can easily be detected. Furthermore, average type data as obtained using the chromatographic methods can also be obtained without difficulties.

NOMENCLATURE

B,	rotational constant;
c,	velocity of light;
C,	constant;
h,	Planck's constant;
I,	intensity;
J,	rotational quantum number;
k,	Boltzmann's constant;
N,	number density;
T,	temperature.

Greek symbols

α,	isotropic part of the polarizability;
γ,	anisotropic part of the polarizability;
μ,	reduced mass of molecule;
ν,	wave number.

Subscripts

A,	anti-Stokes;
S,	Stokes;
0,	incident or initial.

1. INTRODUCTION

MOLECULAR scattering of light in a transparent medium has been known and studied for a long time. In this context one can distinguish between two kinds of scattering. The first, occurring without a change in wavelength of the scattered radiation is known as Rayleigh or Mie scattering depending on the ratio of the diameter of the scatterer to the wavelength of the scattered radiation; the second, accompanied by a change in wavelength of the scattered radiation is known as Raman scattering. The scattering phenomenon accompanied by a wavelength shift was predicted theoretically by Smekal in 1923 and observed experimentally in 1928 by Raman and Krishnan in liquids and by Landsberg and Mandelstam in crystals. Soon afterwards it was also observed in vapors. The importance of these observations was immediately recognized and the development of this new tool for studying molecular properties was very rapid. While molecular and atomic spectroscopy has been known for a long time and applied in physics and chemistry, the special features of the Raman effect makes the diagnostic techniques based upon it of particular value in a number of fields of science and technology. These range from the field of aerospace sciences to the field of air pollution measurements and monitoring, from the field of concentration measurements of combustion products to the field of temperature measurements of jet exhausts. The applications are too many to enumerate. Sufficient to say that wherever there is a need of molecular specie concentration, temperature, relaxation time measurement, etc., the Raman technique can be applied, provided the specie in question is Raman active. The last essentially eliminates all monoatomic species, which as is well-known, are not Raman active in the usual sense.

Since it is almost always of utmost importance in flow field studies, be they of a steady state or transient nature, to obtain a measurement of the local density and in a number of cases to obtain local concentrations of particular species, the Raman diagnostic technique provides a unique method of achieving both without the use of flow disturbing probes. In addition, it provides also the means of obtaining the temperature of the specie of interest.

In this work, after a short review of the Raman scattering theory, the simultaneous determination of

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specie concentration and temperature using Raman scattering is discussed. The necessary apparatus is described. Some of the difficulties encountered in applying this diagnostic technique, based on the work conducted in our laboratory, are discussed.

It is shown that this technique permits the simultaneous measurement of the concentration of several species of a gas mixture, irrespective of its energy state and the respective temperatures of the component species in the gas mixture. The unique ability of this technique to yield the above is made more significant by the fact that only one primary illuminating source is necessary, and the data are obtained remotely. The number of spacially distributed points obtainable simultaneously in the flow field are limited by the optical resolution and the available receiving stations.

Experimental data obtained in a flow field of a turbulent CO_2–air jet and a methane air–CO_2 enriched burner are presented. The results pertaining to the concentration distribution are compared with data obtained utilizing standard chromatographic sampling methods. It is shown that while the standard chromatographic methods are capable of measuring mean values of concentration of a particular specie in a flow field limited to a mixture of two different species, the Raman scattering technique is capable of resolving any number of species in a gas mixture simultaneously. Furthermore, it is shown that the Raman scattering technique is capable of yielding simultaneously with the concentration, the temperature of the given specie, which the standard diagnostic methods are uncapable of providing. Finally, it is shown that the Raman scattering technique, due to its instantaneous response, is capable of resolving fluctuations in density and temperature at a point in the flow field without disturbing probes.

2. THE RAMAN EFFECT

The Raman effect relates essentially to a scattering phenomenon of electromagnetic radiation which is shifted in frequency from primary radiation, and this frequency shift is governed by the character of the scattering specie. The Raman effect has been described in many books and scientific articles [1–13] too numerous to list.

As a result of the wave mechanical approach of Smekal and the subsequent polarizability theory interpretation by Placzek, it can be shown that the Raman scattered intensity can be expressed for vertically polarized primary illumination and normal observation as

$$I_{s_A} = \frac{2^7\pi^5}{135}\frac{hnI_0}{8\pi^2 c\mu v}\frac{(\nu_0 \mp \nu)^4(45\alpha'^2 + 7x\gamma'^2)}{1-\exp\left(\dfrac{-hc\nu}{kT}\right)} \tag{1}$$

where α' and γ' are the change of the isotropic and anisotropic parts of the polarizability in fluids.

The relative intensities of the Stokes and anti-Stokes lines may be expressed by

$$\frac{I_s}{I_A} = \frac{(\nu_0-\nu)^4}{(\nu_0+\nu)^4}\exp\left(\frac{hc\nu}{kT}\right) \tag{2}$$

and

$$T = hc\nu/k\left[\ln\frac{I_s}{I_A} + 4\ln\left(\frac{\nu_0+\nu}{\nu_0-\nu}\right)\right]. \tag{3}$$

It is, therefore, clear that equations (1) and (3) are sufficient to obtain both the number density and the temperature of any molecular specie in a gas.

A close inspection of equation (2) and its plot in Fig. 1 reveals that the Stokes to anti-Stokes Raman scattered intensity ratio method of obtaining temperatures of a species at low or near ambient atmospheric temperatures might present some difficulties due to the very small anti-Stokes intensities. However, it is well-known that the pure rotational Raman spectra appearing near the exciting radiation frequency can be quite intense.

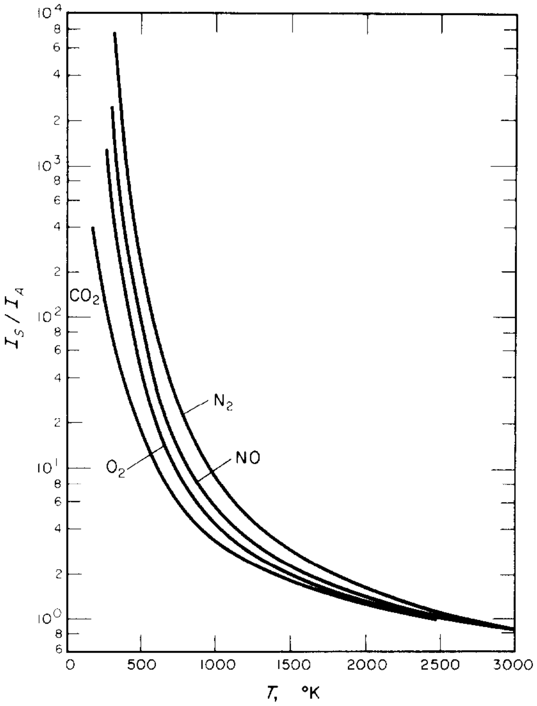

FIG. 1. Ratio of the Stokes to anti-Stokes intensity as a function of gas temperature.

The intensity of the rotational Stokes and anti-Stokes lines can be expressed as

$$I_{sJ} = CN(J+1)(J+2) \times\left[(\nu_0 - 4B(J+3/2))^4\exp(-BJ(J+L))\frac{hc}{kT}\right] \tag{4}$$

$$I_{AJ} = CN(J+1)(J+2) \times \left[(\nu_0+4B(J+3/2))^4 \exp(-B(J+2)(J+3))\frac{hc}{kT}\right] \quad (5)$$

and the ratio as

$$\frac{I_{sJ}}{I_{AJ}} = \frac{[\nu_0-4B(J+3/2)]^4}{[\nu_0+4B(J+3/2)]^4}\exp 4B(J+3/2)\frac{hc}{kT}. \quad (6)$$

From the last equation the temperature is given by

$$T = \frac{4B(J+3/2)hc}{k\left\{\ln\frac{I_s}{I_A}+4\ln\left[\frac{\nu_0+4B(J+3/2)}{\nu_0-4B(J+3/2)}\right]\right\}}. \quad (7)$$

An inspection of equations (4) and (5) reveals that in principle the temperature can be obtained also by taking a ratio of two Stokes or two anti-Stokes lines of different quantum numbers J. Some of the features are shown in Figs. 2 and 3.

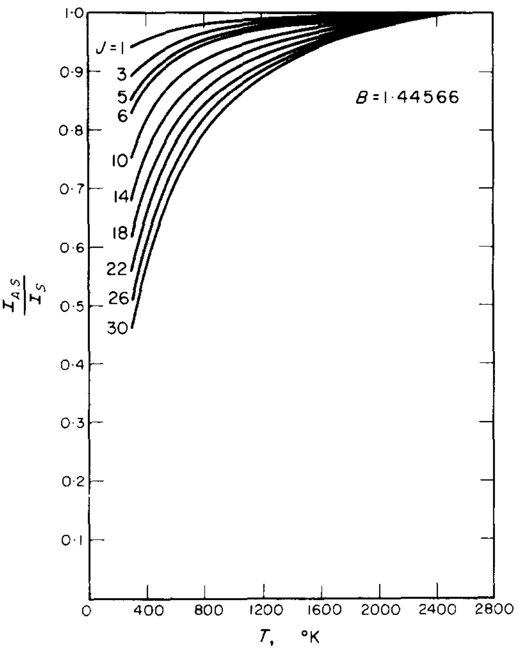

FIG. 2. Rotational anti-Stokes to Stokes intensity as a function of temperature with J as a parameter.

The aim of this work is the demonstration of the ability of obtaining the number density and temperature of a specie in a mixture of gases instantaneously and simultaneously. From equations (1) and (3) it is evident that these variables can be formally obtained from a measurement of the scattered intensity.

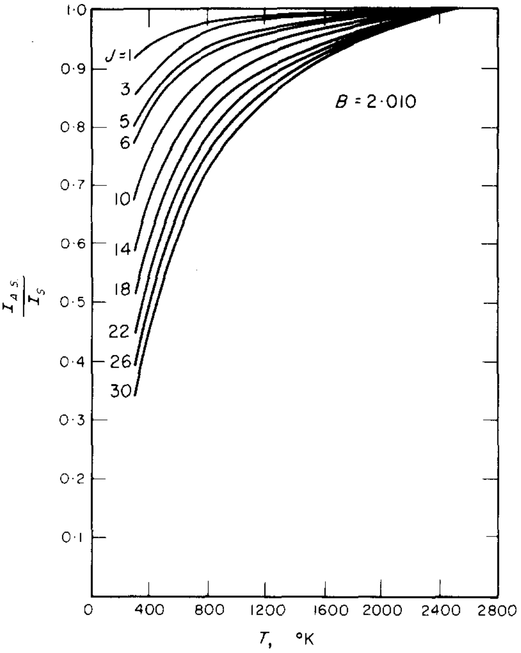

FIG. 3. Rotational anti-Stokes to Stokes intensity as a function of temperature with J as a parameter.

3. EXPERIMENTAL DESIGN CONSIDERATIONS

As indicated above, instantaneous and simultaneous data for the determination of specie concentration and temperature can formally be obtained, the former because the Raman transition takes place in a time of the order of 10^{-14} of a second for the visible range of primary illumination, and the second because one may record the Stokes and anti-Stokes intensity at the same time, the number of data points depending on the number of receiving channels one has available. An obvious difficulty in performing Raman intensity measurements is the extremely small equivalent Raman scattering cross-section. The cross-section defined as the ratio of the incident to the scattered intensity is a function not only of the molecular invariants and characteristic frequency shifts but is strongly affected by the incident frequency as is evident from equation (1). Typically this cross-section may vary between 10^{-28} cm^2 and 10^{-30} cm^2 depending on the molecule under investigation and the frequency of the primary light. Since the frequency dependence is of overriding importance here (fourth power), and is essentially the only parameter which is at the disposal of the experimentalist, one would tend to automatically choose the laser operating at the highest frequency. While this choice might be desirable in one respect, other aspects of this choice might be less favorable. As is well-known, the two most favored of lasers considered for the Raman scattering diagnostic purpose are the ruby laser (6943 Å) and the nitrogen laser (3371 Å). From the purely frequency point-of-view, the scattered intensity resulting from a nitrogen laser is approximately eighteen times higher per unit primary power than that resulting from a ruby laser. While the available power per pulse of a nitrogen laser is of the order of

100 kW, the power per pulse of a ruby laser is of the order of 100 MW with higher power units readily available. On the other hand, nitrogen lasers of several hundred pulses per second are possible while ruby lasers of only 1 pulse per second are available.

Another aspect of Raman scattering must be considered in choosing a laser. The line separation of the Raman shifted radiation is better from the long wavelength primary radiation than from the shorter wavelength radiation. This can be seen in Table 1 where a comparison is made for several species of interest, illuminated with a ruby and a nitrogen laser, respectively. This feature may become important in cases where several species are involved and their measurement and resolution is desired, in particular when narrow bandpass filters are contemplated. It should also be pointed out that the use of a nitrogen laser requires the utilization of photon counting techniques and generally mean values of the measured variables are obtained, while with a ruby laser, due to its much higher power per pulse (several orders of magnitude), single pulse operation is possible and therefore instantaneous values can be obtained. While the problem of band elimination filters is much more complicated for the nitrogen laser than for the ruby laser, the eye safety problems are more severe for ruby lasers than for the nitrogen lasers. Thus, the choice of a laser for diagnostic purposes depends largely on the experiment to be performed and on the balance of the pro and con features of both.

Table 1. Raman wavelength of some molecules corresponding to ruby and N_2 lasers as primary illuminators

	λ_s[Å] Ruby laser	λ_s N_2 laser
NO	7982·5	3597·8
SO_2	7546·2	3506·4
	7201·8	3430·2
	7668·0	3532·5
NO_2	7643·3	3527·2
	7324·2	3457·7
	7821·4	3564·7
H_2O	9305·5	3844·1
	7807·4	3561·8
	9392·1	3858·8
CO_2	7683·5	3535·8
	7623·4	3523·0
CO	8156·8	3632·8
N_2	8283·4	3657·6
O_2	7784·4	3556·9

4. DESIGN OF THE EXPERIMENT

As indicated above, the aim of this work was the demonstration of the applicability of the Raman effect to the determination of number density concentrations of molecular species in a flow field as well as the determination of the temperature of the given specie in the flow field. It was demonstrated [11–12] that both of the above variables can be obtained using the Raman scattering technique in stationary mixtures of gases.

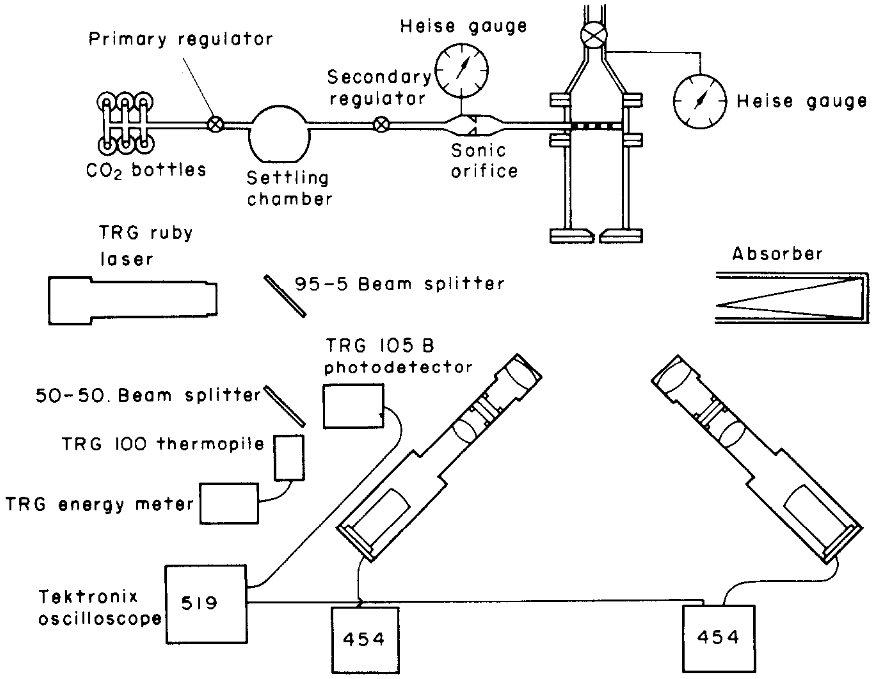

FIG. 4. Schematic diagram of the concentration measurement system.

Having established the feasibility of measurement of concentration in a quiescent mixture of gases, an attempt was made to measure the instantaneous concentration and temperature in a turbulent flow. For that purpose an axisymmetric turbulent CO_2–air jet was utilized. The complete experimental facility is shown schematically in Fig. 4 and photographically in Fig. 5. As can be seen from the above figures, it consists of a ruby laser with the necessary monitoring and control optics. The receiving optics consisted of two focussed telescopes equipped with the appropriate filters and phototubes. These were adjustable to facilitate profile measurements since the jet and the ruby laser were fixed.

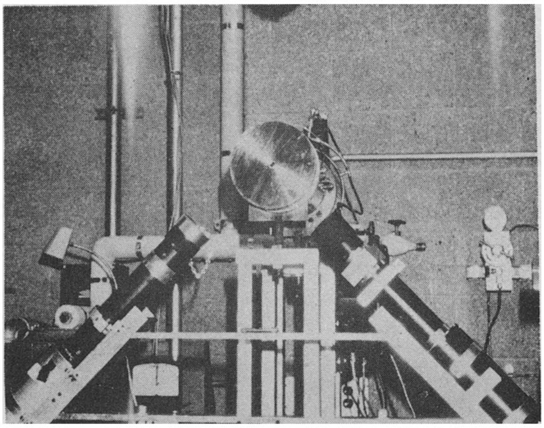

FIG. 5. Photographic view of the experimental apparatus.

While this jet was equipped with the necessary components to provide a heated gas flow, the temperature was limited to about 200°F. It was, therefore, decided to substitute a methane–CO_2 enriched air burner for the jet for the temperature measurements. This decision was based on the results of equation (2) plotted in Fig. 1. It was desired to have Stokes and anti-Stokes scattered intensities of the same order of magnitude. Figure 6 indicates the schematic diagram of the experimental apparatus utilized to measure the temperature. As can be noted, a double monochrometer was used here to measure the anti-Stokes line intensity and a narrow bandpass filter for the Stokes line intensity.

5. EXPERIMENTAL RESULTS AND DISCUSSION

Figures 7 and 8 present some experimental results of the CO_2 concentrations in jets. Included in these figures are the mean values of CO_2 concentration as obtained using the conventional chromatographic methods [14]. Figure 9 presents some preliminary normalized temperatures as obtained from the ratio of Stokes to anti-Stokes intensity measurements. The above concen-

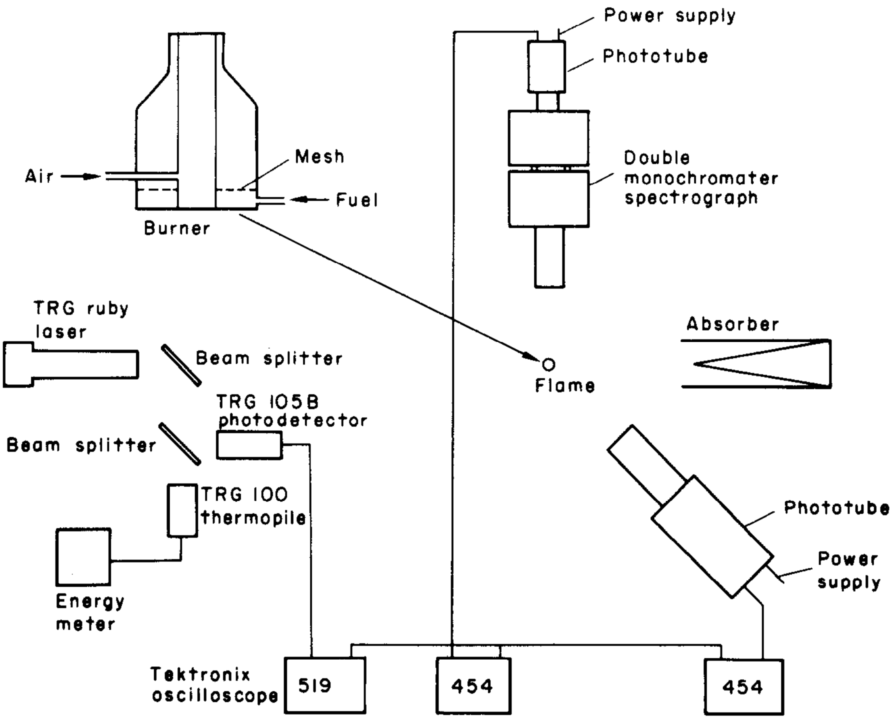

FIG. 6. Schematic diagram of the temperature measurement system.

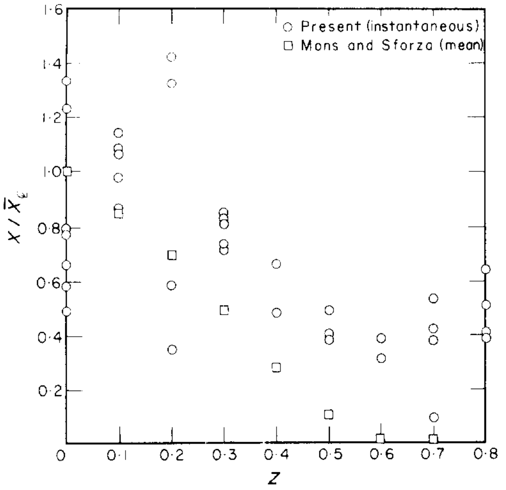

FIG. 7. Instantaneous specie concentration of CO_2 in a turbulent jet at $x/D = 5$.

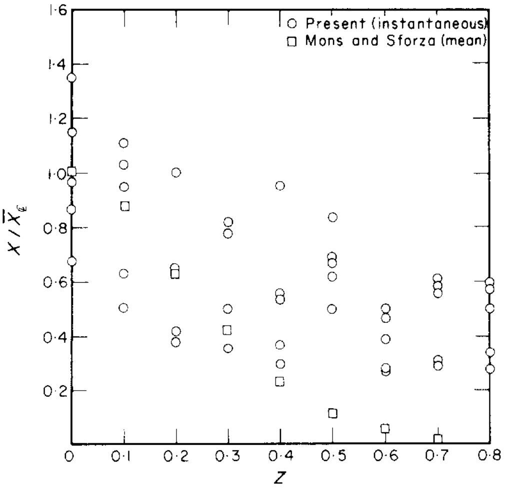

FIG. 8. Instantaneous specie concentration of CO_2 in a turbulent jet at $x/D = 10$.

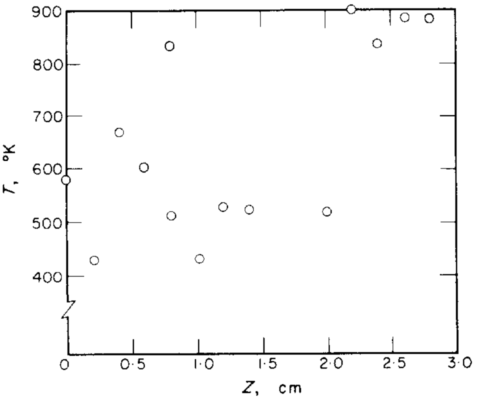

FIG. 9. Temperature distribution.

tration and temperature measurements are, as indicated previously, instantaneous measurement. An inspection of Figs. 7 and 8 might, at a first glance, suggest that the scatter of the data is extremely high if one compares it to the mean profiles as obtained from the chromatographic sampling probes. However, if one considers that the Raman data represent instantaneous values at a given point, one can see that any fluctuation in concentration, temperature or other flow field variables measured cannot be observed by the averaging process necessary, for example, in conductivity cell sampling methods. It should be noted that at each station several measurements were made at random. It is possible to obtain from these measurements mean values similar to those obtained using the conductivity cell sampling method, without the use of probes. However, for meaningful statistical data, a large number of measurements at a point are necessary. Comparing the concentration data obtained in the jet, Figs. 7 and 8, with those obtained in a stationary mixture of gases, Fig. 10, one immediately sees that what appears to be a random scatter of the data is, in reality, an indication of the turbulent fluctuation of the concentration and temperature in the jet or flame. It must be pointed out here that while the short duration pulsed laser is sufficient to indicate turbulent fluctuations of the flow field, a time history of the same is only possible with a continuous laser or pulsed laser where the pulse rate is higher than the turbulent fluctuations of interest.

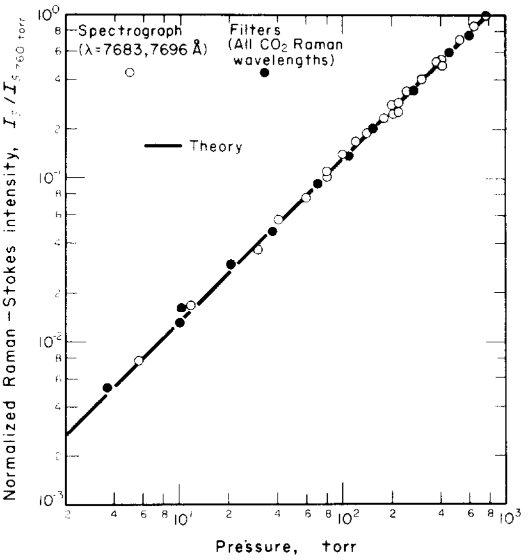

FIG. 10. Measured Raman–Stokes intensity of carbon dioxide (CO_2) as a function of pressure measured in air at room temperature.

Since continuous wave lasers of the proper frequency and power are not readily available, time resolved turbulent fluctuations in specie concentration or temperature by means of Raman scattering is, at present, very difficult to achieve. However, velocity fluctuations, utilizing the Doppler effect, are readily achievable with the available low power C–W lasers. Further developments in laser technology and measurement techniques as well as in the field of resonant Raman techniques should result in improving the flow field diagnostics.

As indicated above, it is possible using the single pulse technique to obtain mean values of the concentration, as well as temperature profiles. Since the system as shown in the schematic diagram, Fig. 4, utilized so far in this work, consisted of a ruby laser capable of delivering one laser pulse in 3 min, the data as shown were obtained using high speed oscilloscopes and polaroid film cameras.

A new system capable of providing both single pulse as well as mean values of the given data has been designed. A schematic diagram of this new system is shown in Fig. 11. It consists of a ruby laser capable of delivering a 150 MW-20 nanosecond duration Q-switched laser pulse at a repetition rate of 6 ppm. The data acquisition was accomplished using preamplifiers, linear gates, and stretching modules, analog to digital converters and an on-line digital computer and teleprinter output. This system permitted not only the acquisition of mean values of a turbulent flow, but also the retrieval of data corresponding to single pulse operation. Thus instantaneous values at a given point could always be obtained. Some preliminary results are shown in Fig. 12. The mean concentration profiles shown here are a result of averaging over fifty individual pulses. As can be seen, the mean values tend to approach the mean values as obtained using the chromatographic methods mentioned earlier.

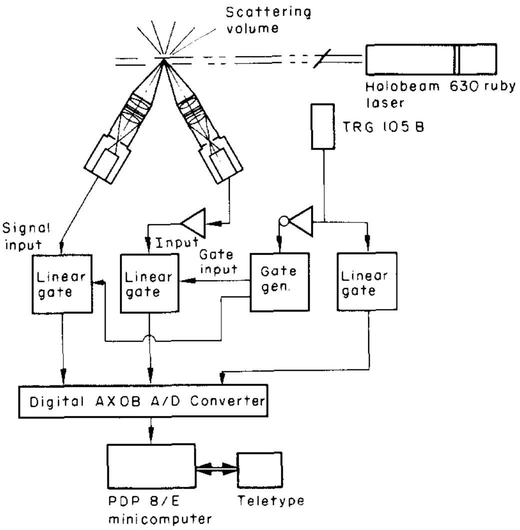

FIG. 11. Schematic diagram of the experimental configuration.

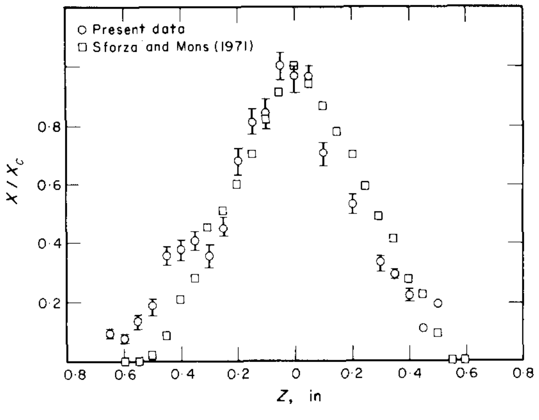

FIG. 12. Specie concentration profile in turbulent jet at $x/D = 5{\cdot}0$.

Acknowledgement—This research was sponsored by the Office of Naval Research and is administered by Purdue University through Contract No. N00014-67-A-0005, NR-098-038, Subcontract No. 4965-36.

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MESURE DE LA TEMPERATURE ET DE LA CONCENTRATION EN ESPECE DANS UN ECOULEMENT

Résumé La technique de la diffusion Raman et son application au diagnostique des champs d'écoulement sont discutées. On montre expérimentalement que la technique est capable de fournir la concentration en espèce et la température du mélange de gaz polyatomiques, non seulement dans le cas du repos mais aussi dans un écoulement. Du fait de la transition Raman extrêmement rapide, en utilisant la technique simple du laser pulsé à haute puissance, on peut détecter aisément les fluctuations de concentration d'espèce et de température. De plus les valeurs moyennes, comme on les obtient par les méthodes chromatographiques, peuvent être mesurées sans difficulté.

MESSUNG VON TEMPERATUR UND KONZENTRATION IM STRÖMUNGSFELD

Zusammenfassung—Die Ramon-Spektroskopie und ihre Anwendung bei der Untersuchung von Strömungsfeldern wird erläutert. Es wird experimentell nachgewiesen, daß diese Technik Aufschluß über Konzentration und Temperatur nicht nur in ruhenden Gemischen mehratomiger Gase, sondern auch im Strömungsfeld vermittelt. Ferner wird gezeigt, daß mit extrem starker Ramon-Durchstrahlung bei Anwendung der Impuls-Laser-Technik Schwankungen von Temperatur und Konzentration im Strömungsfeld leicht nachgewiesen werden können. Darüberhinaus können Mittelwerte wie bei der Anwendung der Chromatographie gewonnen werden.

ИЗМЕРЕНИЯ ТЕМПЕРАТУРЫ И КОНЦЕНТРАЦИИ КОМПОНЕНТОВ В ПОТОКЕ

Аннотация — Обсуждается методика с использованием эффекта рассеивания Рамана и её применение для определения полей в потоке. Экспериментально показано, что использование данного эффекта позволяет определить концентрацию компонентов и значение температуры не только в неподвижной смеси многоатомных газов, но также и в потоке. Показано также, что благодаря большой скорости перехода в этом эффекте с помощью простой импульсной лазерной установки большой мощности можно легко определить существование быстрых флуктуаций концентрации компонентов и температуры в потоке. Кроме того, хроматографическим методом можно также легко получить усредненные значения.