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INVESTIGATION OF LASER RESONANT ABSORPTION FOR GAS DYNAMICS MEASUREMENTS

Frederick M. Shofner and Carl S. Helrich
University of Tennessee Space Institute

August 1971

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FOREWORD

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This technical report has been reviewed and is approved.

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ABSTRACT

A feasibility study of a technique to measure gas densities using resonant scattering of laser light is described. The technique involves the illumination of the gas by a laser beam whose frequency is matched to a natural transition frequency of one species of the gas. The reemitted light is then detected. The density is obtained from the integrated absorption coefficient which is determined by sweeping the laser frequency over the line. A theoretical description is given of line broadening mechanisms. Experimental studies were initiated involving both absorption and reemission of coherent radiation.

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SECTION I

INTRODUCTION

Spectroscopic techniques are attractive in gas dynamic diagnostics. Indeed, under conditions of extremely high temperatures as, for example, in MHD and thermonuclear plasmas, these techniques become the principle ones available. Spectroscopic studies can reveal gas temperatures through measurements of Doppler half-widths, velocities through Doppler shifts, states of ionization through asymmetries and Stark profiles, pressures through collision halfwidths, and densities of various species through integrated absorption coefficients. It is clear that in principle most physical properties of the medium containing the radiating atoms are somehow reflected in the line structure. Once a fuller understanding of how these properties affect the line structures is gained and further experimental techniques are developed, we shall have at our disposal a non-interfering probe which should prove valuable in gasdynamic diagnostics.

Before the appearance of the laser, spectroscopic techniques were, for the most part, limited to studies of emission spectra from hot gases or the study of absorption of a white light source. The laser, however, constitutes a source of extremely narrow bandwidth and the inevitable further development of tunable lasers will provide us with a very selective probe with which we can study the structure of an individual line of a given species. By judiciously choosing the laser frequency, the laser beam will interact with only a certain set of atoms capable of undergoing a known transition whose wavelength matches that of the laser. By sweeping the laser frequency over the broadened line, the absorption coefficient $k(\nu)$ may be measured directly. With an absorption experiment, $k(\nu)$ may be time resolved. With an emission experiment, $k(\nu)$ may be both time and space resolved.

The effort described in this report was directed toward obtaining a measurement of the density of atomic oxygen in a hypersonic wind tunnel. The original idea that resonant absorption could be used to obtain this measurement grew out of a February 1967 meeting between Dr. H. K. Doetsch, M. K. Kingery (AEDC) and Dr. F. M. Shofner (UTSI) in which the interesting possibilities of resonant absorption were discussed. At low densities the absorption coefficient is directly proportional to the one particle distribution function of kinetic theory from which one can calculate density. This conclusion has been confirmed experimentally, using the laser by others (1,2,3) and by the UTSI group.

However, the necessary conditions for this simple propor-

tionality are not satisfied in the hostile environment to be encountered in the hypersonic, arc-driven wind tunnel. The processes contributing to the shape of the spectral line are very complicated and the shape may be quite different from the simple Gaussian of the low density regime. Consequently, the theoretical basis of the research presented here has shifted from simple kinetic theory to basic radiation theory. The result is, however, quite similar. The density is still to be determined from a measurement of the integrated absorption coefficient.

Section II of this report deals with a presentation of the basic broadening mechanisms and their individual and combined effects on the line shape. Section III describes the apparatus used in the experimental portions of the work and the experiments conducted. Section IV gives a review of the results together with an analysis of the feasibility of the application of resonant absorption to measure gas densities.

SECTION II

THEORETICAL CONSIDERATIONS

A spectral line may be broadened by three basic mechanisms: the finite lifetime of the state resulting in an uncertainty of the energy of the state and, therefore, the emission frequency, the Doppler effect resulting from thermal motion of the atoms or molecules, and collisional effects which cause shifts in the energy states. Natural broadening and collisional broadening produce a Lorentzian profile while the Doppler broadened profile is Gaussian. The general spectral line from an assembly of gas atoms possessing thermal energy will be a combination of both of these profiles.

The effects of these broadening mechanisms are buried in the phenomenological absorption coefficient, $k(\nu)$, defined by the relation.

$$dI_{\text{abs}}(\nu)d\nu = -I(x,\nu) k(\nu) d\nu dx \quad (1)$$

where

$dI_{\text{abs}}(\nu)d\nu$ = light intensity absorbed in the frequency interval $(\nu \rightarrow \nu + d\nu)$ * and in the spatial element $(x \rightarrow x + dx)$ * along the beam.

$I(x,\nu)d\nu$ = light intensity in the frequency interval $(\nu, d\nu)$ at the point x .
 $I(x,\nu)$ is referred to as the specific intensity.

A general expression for $k(\nu)$ can be derived by relying on basic radiation theory.

Consider a transition between a lower quantum state (l) and an upper quantum state (u) with an energy $h\nu_{lu}$. ν_{lu} is then the natural frequency of the transition. Let there be n_l atoms (molecules) per unit volume in the state (l). Owing to the presence of perturbers and thermal motion of the atoms, only $n_l(\nu)d\nu$ atoms per unit volume are capable of absorbing light in the frequency interval $(\nu, d\nu)$. According to Einstein's theory of radiation (4), the number of photons absorbed in the

*Throughout the remainder of this report we shall refer to such an interval as $(\nu, d\nu)$ or (x, dx)

interval $(\nu, d\nu)$ per unit time per unit volume from a beam of specific intensity $I(\nu)$ is

$$\frac{1}{4\pi} n_l(\nu) d\nu B_{lu} I(\nu) \quad (2)$$

and number of photons stimulated to emit into the beam per unit time per unit volume is

$$\frac{1}{4\pi} n_u(\nu) d\nu B_{ul} I(\nu) \quad (3)$$

where

B_{lu} = Einstein absorption coefficient

B_{ul} = Einstein stimulated emission coefficient

In addition, Einstein introduced a spontaneous emission coefficient, A_{ul} , and proved the relations (4,5)

$$\frac{A_{ul}}{B_{lu}} = \frac{g_l}{g_u} \frac{2h\nu^3}{c^2} \quad (4a)$$

$$g_l B_{lu} = g_u B_{ul} \quad (4b)$$

where g_l and g_u are the statistical weights of the states (l) and (u) respectively. Furthermore, it should be mentioned that the Einstein coefficients are functions of atomic parameters only and are independent of statistical parameters (e.g. temperature, pressure, and density) (4,5).

Let a light (laser) beam of cross sectional area dA be directed along the x -axis. If the specific intensity of the beam at the point x is $I(x, \nu)$, the net amount of energy lost from the beam per unit time in a distance (x, dx) in the interval $(\nu, d\nu)$ is from Equation (1)

$$-dI(\nu) d\nu dA = I(x, \nu) d\nu k(\nu) dA dx \quad (5a)$$

The net number of photons absorbed per unit time is then

$$\frac{1}{h\nu} I(x, \nu) d\nu k(\nu) dA dx \quad (5b)$$

From Equations (2) and (3) the net number of photons absorbed per unit time in a volume $dA dx$ is

$$\frac{1}{4\pi} I(x, \nu) d\nu [n_l(\nu) B_{lu} - n_u(\nu) B_{ul}] \quad (6)$$

Equating (5b) and (6), we have

$$\frac{1}{h\nu} k(\nu)d\nu = \frac{1}{4\pi} [n_\ell(\nu) B_{\ell u} - n_u(\nu) B_{u\ell}]d\nu \quad (7)$$

Much of the literature appearing today is written in terms of the normalized lineshape function, $g(\nu)$. This function is defined by writing

$$n_\ell(\nu) = N_\ell g(\nu) \quad (8)$$

where

$$N_\ell = \int d\nu n_\ell(\nu) \quad (8a)$$

is the total number of particles in the state ℓ . $g(\nu)$ then contains information regarding the physical mechanisms broadening the line.

With this, Equation (7) may be written

$$\frac{1}{h\nu} k(\nu)d\nu = \frac{1}{4\pi} [N_\ell B_{\ell u} - N_u B_{u\ell}]g(\nu)d\nu. \quad (9)$$

In deriving Equation (9) it has been assumed that

$$n_u(\nu) = N_u g(\nu). \quad (9a)$$

The validity of this assumption can be argued from the fact that $n_\ell(\nu)$ and $n_u(\nu)$ both depend on the separation between energy states ($E_u - E_\ell$) and not on the states themselves. The separation of the states is a function of the eigenstate energies and statistical parameters such as temperature, pressure and degree of ionization. While the individual states may be affected differently by these statistical parameters, the assumption is that the state difference, which is a combination of both effects, is ultimately a function only of statistical parameters and is the same for absorption or emission.

Using (4), Equation (9) becomes

$$\frac{1}{h\nu} k(\nu)d\nu = \frac{1}{8\pi} \frac{1}{h\nu} \frac{c^2}{\nu^2} A_{u\ell} \left[N_\ell \frac{g_u}{g_\ell} - N_u \right] g(\nu)d\nu \quad (10)$$

The mean lifetime of the state (u) is defined by

$$\tau = \frac{1}{A_{u\ell}} \quad (11)$$

With this, Equation (10) becomes

$$\frac{1}{h\nu} k(\nu) d\nu = \frac{1}{8\pi} \frac{1}{h\nu} \frac{\lambda^2}{\tau} N_l \frac{g_u}{g_l} \left[1 - \frac{g_l}{g_u} \frac{N_u}{N_l} \right] g(\nu) d\nu \quad (12)$$

where $\lambda (= \frac{c}{\nu})$ is the wavelength of the transition.

The absorption coefficient is then

$$k(\nu) = \frac{1}{8\pi} \frac{\lambda^2}{\tau} N_l \frac{g_u}{g_l} \left[1 - \frac{g_l}{g_u} \frac{N_u}{N_l} \right] g(\nu) \quad (13)$$

Assuming that the line width $\Delta\nu$ satisfies

$$\Delta\nu \ll \nu,$$

the factor $\lambda^2 = c^2/\nu^2$ may be assumed to be slowly varying over the line width. Equation (13) may be integrated to give

$$\begin{aligned} \int_{-\infty}^{+\infty} k(\nu) d\nu &= \frac{1}{8\pi} \frac{\lambda_{lu}^2}{\tau} N_l \frac{g_u}{g_l} \left[1 - \frac{g_l}{g_u} \frac{N_u}{N_l} \right] \int_{-\infty}^{+\infty} g(\nu) d\nu \\ &= \frac{1}{8\pi} \frac{\lambda_{lu}^2}{\tau} N_l \frac{g_u}{g_l} \left[1 - \frac{g_l}{g_u} \frac{N_u}{N_l} \right] \end{aligned} \quad (14)$$

It should be noted that in the derivation of (14) no assumption was made regarding the broadening mechanism. Therefore, (14) holds regardless of the environment in which the atoms find themselves.

If $N_u \ll N_l$, a situation usually realized when N_u is populated mainly by the beam, Equation (14) becomes

$$\int_{-\infty}^{+\infty} k(\nu) d\nu = \frac{\lambda_{lu}^2}{8\pi} \frac{N_l}{\tau} \frac{g_u}{g_l} \quad (15)$$

This states that the integrated absorption coefficient is directly proportional to the density of absorbers independent of all other occurrences.

In general, the ratio N_l/N_u can be determined from a knowledge of the density matrix. In equilibrium the atoms are distributed according to the so-called Boltzmann factor and the ratio N_l/N_u can be calculated simply. We shall now discuss Equation (15) for various broadening mechanisms.

NATURAL BROADENING

So-called natural line broadening is related to the Heisenberg uncertainty principle which may be stated in the form

$$\Delta E \Delta t \gtrsim \frac{1}{2} \hbar \quad (16)$$

Since the lifetime of the state, with which we may identify Δt , is finite, the uncertainty in state energy, ΔE , is non-zero. This uncertainty in state energy produces an uncertainty in the transition frequency given by

$$\Delta \nu_{\ell u} = \Delta \nu_{\ell u} = \frac{1}{h} (\Delta E_u + \Delta E_\ell) = \frac{1}{2} \pi \Delta \nu_n \quad (17)$$

where ΔE_u and ΔE_ℓ are the uncertainties in the energies of the upper and lower states and $\Delta \nu_n$ is the natural half-width (the width of the profile at half maximum). The quantitative treatment of natural broadening is based on the Dirac theory of radiation (6,7,8). The results of such a treatment show that the normalized time shape function for the naturally broadened line is of the form

$$g_n(\nu - \nu_{u\ell}) = \frac{\Delta \nu_n}{4\pi^2 (\nu - \nu_{u\ell})^2 + \frac{1}{4} (\Delta \nu_n)^2} \quad (18)$$

Such a profile is termed Lorentzian. Calculations show that the natural halfwidth is extremely small and will be neglected hereafter.

DOPPLER BROADENING

To consider pure Doppler broadening, we shall ignore all interactions. This is equivalent to assuming a low density and no perturbing external fields.

If an atom with a natural frequency $\nu_{\ell u}$ is moving along the beam axis with a velocity v_x , then the frequency absorbed (or emitted) by the atom as measured in the laboratory frame is

$$\nu = \nu_{\ell u} \left(1 - \frac{v_x}{c} \right) \quad (19)$$

If the one-particle distribution function is $f(\underline{r}, \underline{v}, t)$, the probability that an atom in the volume element $(\underline{r}, d\underline{r})$ will have an x-velocity in the range (v_x, dv_x) at the time t is

$$d\underline{r} dv_x \int_{-\infty}^{+\infty} dv_y \int_{-\infty}^{+\infty} dv_z f(\underline{r}, \underline{v}, t) \quad (20)$$

Using (19) in (20), one has for the probability that an atom in $(\underline{r}, d\underline{r})$ will absorb a photon in the range $(|v - v_{lu}|, dv)$ the expression

$$d\underline{r} \left(\frac{c}{v_{lu}} \right) d(|v - v_{lu}|) \int_{-\infty}^{+\infty} dv_y \int_{-\infty}^{+\infty} dv_z f(\underline{r}, v_y, v_z, \frac{c}{v_{lu}} (v - v_{lu}), t) \quad (21)$$

For the case of thermodynamic equilibrium, (21) becomes

$$\left(\frac{Mc^2}{2\pi kT v_{lu}^2} \right)^{\frac{1}{2}} e^{-\frac{M}{2kT} \frac{c^2}{v_{lu}^2} (v - v_{lu})^2} dv \quad (22)$$

where

M = mass of atom

c = speed of light

k = Boltzmann's Constant

T = Absolute temperature of gas.

From here the absorption coefficient defined by Equation (1) becomes

$$k(|v - v_{lu}|) = \frac{c^2}{8\pi v_{lu}^2} A_{ul} N_l \frac{g_u}{g_l} \left(1 - \frac{g_l}{g_u} \frac{N_u}{N_l}\right) \left(\frac{Mc^2}{2\pi kT v_{lu}^2} \right)^{\frac{1}{2}} e^{-\frac{Mc^2}{2kT v_{lu}^2} (v - v_{lu})^2} \quad (23)$$

from which we may identify

$$g_{\text{doppler}}(\nu - \nu_{lu}) = \left(\frac{Mc^2}{2\pi T\nu_{lu}^2} \right)^{\frac{1}{2}} e^{-\frac{Mc^2}{2kT\nu_{lu}^2} (\nu - \nu_{lu})^2} \quad (23a)$$

Since

$$\int_{-\infty}^{+\infty} d\nu \left(\frac{Mc^2}{2\pi kT\nu_{lu}^2} \right)^{\frac{1}{2}} e^{-\frac{Mc^2}{2kT\nu_{lu}^2} (\nu - \nu_{lu})^2} = 1, \quad (24)$$

Equation (12) is easily recovered from Equation (23).

One important conclusion can be drawn from (23). The maximum value of the absorption coefficient

$$k_{\text{max}} = \frac{c^2}{8\pi\nu_{lu}^2} A_{ul} N_l \frac{g_u}{g_l} \left[1 - \frac{g_l}{g_u} \frac{N_u}{N_l} \right] \left(\frac{Mc^2}{2\pi kT\nu_{lu}^2} \right)^{\frac{1}{2}}, \quad (25)$$

found by setting $\nu = \nu_{lu}$, is proportional to the density of atoms in the lower state, N_l , assuming $N_u \ll N_l$. Therefore, in the Doppler regime the density can be determined from a measurement of only k_{max} if the temperature is known. For the case of a rarefied gas, then, a density measurement is possible without sweeping the line. Alternatively, the temperature can be determined from the line width. From Equations (23) and (25), the Doppler half-width may be calculated:

$$\Delta\nu_D = \left(\frac{2 \ln(2) kT\nu_{lu}^2}{Mc^2} \right)^{\frac{1}{2}}$$

COLLISIONAL AND STARK BROADENING

The mechanisms of collisional and Stark broadening involve a shift in the energy levels due to an alteration of the internal field of an atom during collisions with either neutral atoms or ions and electrons. Based on an analysis in which both the atom and perturber are treated quantum mechanically, M. Baranger (9) and M. B. Lewis (10) have shown that under conditions of moderate density the collision or Stark broadened profile is a Lorentzian profile:

$$g_c(\nu - \nu_{lu}) = \frac{\text{Constant}}{(\nu - \nu_{lu})^2 + \left(\frac{\nu_C}{2\pi}\right)^2} \quad (26)$$

where ν_C is the "collision frequency" which, of course, for charged particles is not well defined.

The maximum value of the absorption coefficient in the collision broadened case can be shown to be (8,11)

$$k_{\max} = \frac{\lambda^2}{8\pi} \frac{\nu_{lu}}{\tau\nu_C} \frac{g_u}{g_l} N_l \left(1 - \frac{g_l}{g_u} \frac{N_u}{N_l}\right) \quad (27)$$

For a single species gas, both the collision frequency, ν_C , and the number densities, n_l and n_u , are proportional to the total number density (and hence the pressure). Therefore, (27) is actually independent of number density (pressure). This has been verified experimentally by Gerry and Leonard (11). However, Equation (14) is still valid so that the number density can still be determined from a measurement of the integrated absorption coefficient.

In addition to broadening the line, collisions may also shift the center of the line and produce asymmetries. The shifts may be calculated from quantum mechanical scatter theory. Baranger (9) has shown them to be proportional to the real part of the forward scattering amplitude which, in the case of a spherically symmetric potential (which will, in the shielding approximation, be realized closely by atomic oxygen), is given by (12)

$$\frac{1}{2iK} \sum_l (2l + 1) (e^{2i\delta_l} - 1) P_l(\cos\theta) \quad (28)$$

where

K = wave number of scatterer

δ_l = scattering phase shift

θ = scattering angle

P_l = Legendre Polynomial

Both Baranger (9) and Anderson (13) have indicated schemes by which shifts and shapes of spectral lines can be computed.

Quantum mechanically, the halfwidth of a spectral line is proportional to the imaginary part of the forward scattering amplitude (9,10). However, there are classical and semi-classical theories which provide much simpler results (14, 15, 16). Baranger (9) and Lewis (10) have shown that under conditions comparable to those being considered, the classical impact theory provides suitable results.

An infinitely sharp line implies a radiating process of infinite duration. In impact theory, one assumes that impacts with perturbers shorten the lifetime of the state and thereby broaden the line. In this simple form, impact broadening predicts a quenching of the radiation which has been observed in relatively few cases (15). Therefore, this simple form is not wholly satisfactory. The impact theory has been modified by Lenz and Weisskopf based on the assumption that the effect of a collision is a phase change of the emitted radiation accompanying the detuning of its frequency by a passing particle. If the phase change is large enough, the collision will effectively divide the wave train into two incoherent ones. This is tantamount to termination as far as the line width is concerned.

Calculations based on this theory yield the following expressions for collisional broadened halfwidths (15):

$$\Delta\nu_C \approx 2.2 b^{2/5} (\bar{v})^{3/5} n_1 \quad (29a)$$

for broadening by foreign atoms

$$\Delta\nu_C \approx (e^2 f_{12} / 2\pi M v_{ul}) n_1$$

for broadening by similar atoms

where in (29a)

$$b \approx 10^{-31} - 10^{-32} \text{ cm}^6 \text{ sec}^{-1}$$

$$\bar{v} = \left\{ \frac{8kT}{\pi} \frac{M_1 M_2}{M_1 + M_2} \right\}^{\frac{1}{2}}$$

M_1, n_1 = mass, density of perturbers

M_2 = mass of emitters

and in (29b)

f_{12} = oscillator strength for transition $1 \rightarrow 2$.

COMBINED COLLISIONAL AND DOPPLER BROADENING

Except under controlled conditions (single species, pressure < few Torr), both collision and Doppler broadening will be present. The profile is then a combination of the Doppler (Gaussian) and the collisional (Lorentzian) profiles. To obtain an expression for this general profile, we shall assume that the two broadening mechanisms are independent.

We define the probability that an atom has a characteristic frequency in the interval (v', dv') (resulting from collisional effects) and a velocity in the beam direction (u, du) as

$$L'(v', u) dv' du = L(v', v'') dv' dv'' \quad (30)$$

with

$$v'' = v_{u\ell} \left(1 - \frac{u}{c} \right) \quad (30a)$$

If the mechanisms of Doppler and collisional broadening are statistically independent, this may be expressed as a product of probabilities

$$L(v', v'') dv' dv'' = D(v'' - v_{u\ell}) C(|v' - v''|) dv' dv'' \quad (31)$$

where

$dv'' D(v'' - v_{u\ell})$ = probability that the frequency will be Doppler shifted from $v_{u\ell}$ to v''

$dv' C(v' - v'')$ = probability that the frequency will be shifted from v'' to v' due to collision

From Equation (22) we have

$$D(|v'' - v_{u\ell}|) = \left(\frac{Mc^2}{2\pi kTv_{\ell u}^2} \right)^{\frac{1}{2}} e^{-\frac{Mc^2}{2kTv_{\ell u}^2} (v'' - v_{\ell u})^2} \quad (32)$$

and from Equation (26)

$$\begin{aligned} C(|v' - v''|) &= \frac{\Delta v_C}{2\pi} \frac{1}{(v' - v'')^2 + \frac{(\Delta v_C)^2}{2}} \\ &= \frac{\Delta v_C}{2\pi} \frac{1}{[(v' - v_{\ell u}) - (v'' - v_{\ell u})]^2 + \left(\frac{\Delta v_C}{2}\right)^2} \end{aligned} \quad (33)$$

where Δv_C = collisional halfwidth

Using (32) and (33), (31) becomes

$$\begin{aligned} L(v', v'') dv' dv'' &= \frac{\Delta v_C}{2\pi} \left(\frac{Mc^2}{2kTv_{\ell u}^2} \right)^{\frac{1}{2}} \frac{e^{-\frac{Mc^2}{2kTv_{\ell u}^2} (v'' - v_{\ell u})^2}}{[(v' - v_{\ell u}) - (v'' - v_{\ell u})]^2 + \left(\frac{\Delta v_C}{2}\right)^2} dv' dv'' \end{aligned} \quad (34)$$

The probability that an atom will absorb a frequency in the range (v', dv') is then

$L(v') dv'$

$$= \frac{\Delta v_C}{2\pi} \left(\frac{Mc^2}{2kTv_{lu}^2} \right)^{\frac{1}{2}} dv' \int_{-\infty}^{+\infty} dv'' \frac{e^{-\frac{Mc^2}{2kTv_{lu}^2} (v'' - v_{lu})^2}}{[(v' - v_{lu}) - (v'' - v_{lu})]^2 + \left(\frac{\Delta v_C}{2}\right)^2} \quad (35)$$

From here the absorption coefficient, $k(v)$, is

$$k(v) = \frac{\Delta v_C}{2\pi} \left(\frac{Mc^2}{2kTv_{lu}^2} \right)^{\frac{1}{2}} \int_{-\infty}^{+\infty} dv' \frac{e^{-\frac{Mc^2}{2kTv_{lu}^2} (v' - v_{lu})^2}}{[(v - v_{lu}) - (v' - v_{lu})]^2 + \left(\frac{\Delta v_C}{2}\right)^2} \quad (36)$$

where

$$S \equiv \int_{-\infty}^{+\infty} dv k(v) \quad (36a)$$

Equation (36) may be expressed in a series for small values of the parameter

$$\alpha \equiv \frac{\Delta v_C}{\Delta v_D} \sqrt{\ln 2} \quad (37)$$

Defining

$$\xi \equiv \left(\frac{Mc^2}{2kTv_{lu}^2} \right)^{\frac{1}{2}} (v - v_{lu}) \quad (38a)$$

$$\xi' \equiv \left(\frac{Mc^2}{2kTv_{lu}^2} \right)^{\frac{1}{2}} (v' - v_{lu}) \quad (38b)$$

Equation (36) becomes

$$k(\xi) = \frac{S}{2\pi} \alpha \int_{-\infty}^{+\infty} d\xi' \frac{e^{-\xi'^2}}{(\xi - \xi')^2 + \left(\frac{\alpha}{2}\right)^2} \quad (39)$$

Expanding the integrand for small α , we have

$$\begin{aligned}
k(\xi) &= \frac{S}{2\pi} \lim_{\alpha \rightarrow 0} \alpha \int_{-\infty}^{+\infty} d\xi' \frac{e^{-\xi'^2}}{(\xi - \xi')^2 + \left(\frac{\alpha}{2}\right)^2} \\
&+ \frac{S}{2\pi} \alpha \int_{-\infty}^{+\infty} d\xi' \frac{e^{-\xi'^2}}{(\xi - \xi')^2} + o(\alpha^2)
\end{aligned} \tag{40}$$

We note that as $\xi \rightarrow \xi'$ and $\alpha \rightarrow 0$, the limit in the first term of Equation (40) does not exist in the ordinary sense. However, it can be shown,

$$\begin{aligned}
\lim_{\alpha \rightarrow 0} \frac{1}{\pi} \frac{\alpha/2}{(\xi - \xi')^2 + \left(\frac{\alpha}{2}\right)^2} &= \lim_{b \rightarrow \infty} \frac{1}{\pi} \frac{b}{b^2 (\xi - \xi')^2 + 1} \\
&= \delta(\xi - \xi')
\end{aligned} \tag{41}$$

Therefore

$$\begin{aligned}
k(\xi) &= S \int_{-\infty}^{+\infty} d\xi' \delta(\xi - \xi') e^{-\xi'^2} \\
&+ \frac{S}{2\pi} \alpha \int_{-\infty}^{+\infty} d\xi' \frac{e^{-\xi'^2}}{(\xi - \xi')^2} + o(\alpha^2) \\
&= S e^{-\xi^2} + \frac{S}{2\pi} \alpha \int_{-\infty}^{+\infty} d\xi' \frac{e^{-\xi'^2}}{(\xi - \xi')^2} + o(\alpha^2)
\end{aligned} \tag{42}$$

A general expansion has been given for $k(\xi)$ by Plass and Fivel (8).

$$\begin{aligned}
 k(\xi) = S \{ & e^{-\xi^2} - \alpha \pi^{-\frac{1}{2}} [1 - 2\xi F(\xi)] \\
 & + \left(\frac{\alpha}{2}\right)^2 (1 - 2\xi^2) e^{-\xi^2} \\
 & - 2 \left(\frac{\alpha}{2}\right)^3 \pi^{-\frac{1}{2}} \left[\frac{2}{3} (1 - \xi^2) - 2\xi \left(1 - \frac{2}{3} \xi^2\right) F(\xi) \right] \\
 & + \left(\frac{\alpha}{2}\right)^4 \left(\frac{1}{2} - 2\xi^2 + \frac{2}{3} \xi^4 \right) e^{-\xi^2} + \dots \} \quad (43)
 \end{aligned}$$

Here

$$F(\xi) = e^{-\xi^2} \int_0^\xi e^{\xi^3} d\xi \quad (44a)$$

and

$$F_n(\xi) = \frac{d^n}{d\xi^n} F(\xi) \quad (44b)$$

From Equation (43), one is led to some interesting conclusions. Using Equation (37), we see that as the gas pressure is increased, Δv_C increases and so does α . For small values at $\Delta v_C / \Delta v_D$, however, α remains small so that the term linear in α in Equation (43) is dominant.

Collisional broadening can be due to either atoms of the same species or different species. However

$$S = \int_{-\infty}^{+\infty} dv k(v)$$

is a function only of the number of emitters and absorbers from Equation (14). Therefore, we are led to the conclusion that as we add in foreign atoms to a gas consisting of a fixed density of emitters and absorbers, the integrated absorption coefficient remains constant while the centerline absorption coefficient, $k(\xi = 0)$, decreases. This is seen directly from Equation (43).

One may come to the same conclusion by simpler reasoning as well. Equation (14) implies the constancy of the integrated

absorption coefficients during the addition of foreign atoms to a gas containing a constant number of emitters and absorbers. Because of collisional broadening, however, the profile half-width must increase. The net effect must then be a decrease in the centerline coefficient.

Experiments were performed during the course of this work to test this prediction and to measure the collisional halfwidth for CO_2 being broadened by He. During the experiments, the partial pressure of CO_2 was kept quite low (<5 Torr) so that using Equations (14) and (25)

$$S \approx \left(\frac{2\pi k T v^2 \ell u}{Mc^2} \right)^{\frac{1}{2}} k_{\max}^{(d)}$$

where $k_{\max}^{(d)}$ is the centerline absorption coefficient for pure CO_2 ($P < 5$ Torr). He was then admitted into the test cell holding the partial pressure of CO_2 constant. The centerline absorption coefficient was measured as the partial pressure of He was increased. Using the first three terms of

$$k(\xi = 0) = \left(\frac{2\pi k T v^2 \ell u}{Mc^2} \right)^{\frac{1}{2}} k_{\max}^{(d)} \left\{ 1 - \alpha \pi^{-\frac{1}{2}} + \left(\frac{\alpha}{2} \right)^2 - \frac{4}{3} \left(\frac{\alpha}{2} \right)^3 \pi^{-\frac{1}{2}} + \frac{1}{2} \left(\frac{\alpha}{2} \right)^4 + \dots \right\},$$

α was calculated.

SECTION III

EXPERIMENTAL CONSIDERATIONS

As mentioned previously, spectroscopic techniques involving scattering have been widely applied to MHD and thermonuclear plasmas (References 17 - 22). These techniques involved Q-switched ruby and neodymium Lasers and, more recently, pulsed tunable dye lasers and the interest is centered largely on electron temperatures and density. Also, Karamchetti et al (References 1,2,3) have been able to measure the distribution function of a low pressure ionized gas at rest by a direct measurement of the attenuation of a laser beam as it passed through the absorbing gas. In these experiments, single frequency tunable Helium-Neon laser was used as a light source and a Neon glow discharge as the absorbing medium. However, an extensive literature search has failed to reveal any work done on resonant scattering. Some authors mention the possibility of such measurements (Reference 23), but do not give evidence of any experimental success. It is toward this problem: a demonstration of the feasibility of the use of resonant scattering in gas dynamic diagnostics, that the experimental portion of this work was directed. The choice of experiments performed and apparatus used was dictated by relevance to the stated goal of performing density measurements in a hypersonic wind tunnel.

To obtain pointwise, time-resolved density measurements in a gas flow, the emitted radiation from a small volume common to the illuminating beam and the collector acceptance cone is focused onto the detector. The amount of radiation detected is proportional to the incident laser power and the absorption coefficient at the frequency of the laser light. In this manner, the absorption coefficient may be measured from the re-emitted radiation. The advantage of this procedure over a direct measurement of the absorption coefficient by determining the attenuation of the beam over its length is that it provides a value of the density as a function of x rather than an average density. Thus the ability to measure this re-emitted light became the central problem in this work.

Inherent in the above discussion is the assumption that the laser beam is monochromatic. It is only under such conditions that we can determine the absorption coefficient as a function of frequency. Monochromaticity then became a very important criterion in selecting the laser-absorbing gas system to be used in the study. Also, since only a small percentage of the beam intensity is absorbed in the length dx and only a tiny fraction of that is detected, it was necessary that the laser beam intensity be quite high. Based on these requirements of monochromaticity and high intensity and the fact that the UTSI was already actively involved in work with the CO₂ laser, the CO₂ laser was chosen as the source for the experimental work. The absorbing gas used in

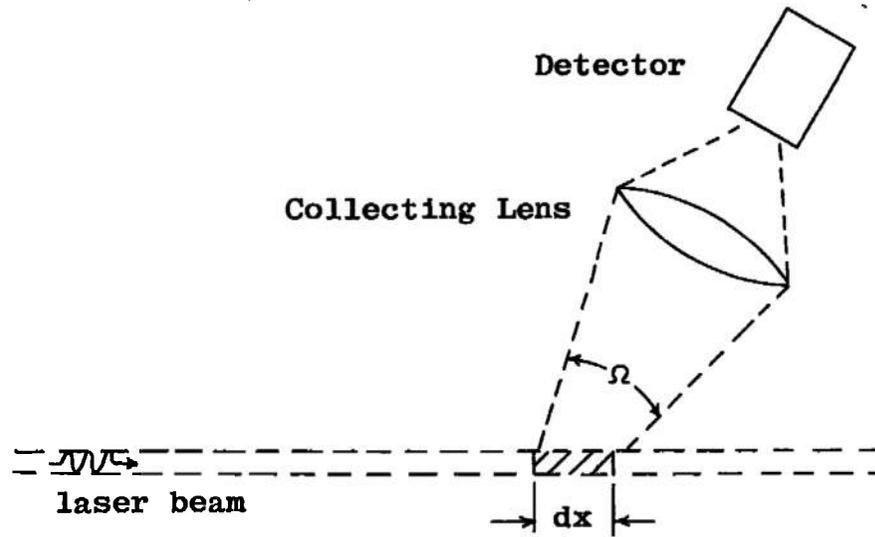


Figure 1. Scattering Schematic

the experiments was also CO_2 . Also very important is the fact that there is a good body of literature on the CO_2 molecule, considerable of it laser-related.

Among the gas systems not considered acceptable for this initial experiment were Helium-Neon and Argon II. Both laser systems were attractive because of the relative simplicity of the visible wavelength detection system. Here one could use a photomultiplier rather than the cryogenically cooled detector necessary for the infrared radiation emitted by the CO_2 lasers. However, the intensity of the Helium-Neon laser would have been low compared to the CO_2 laser and it would have been necessary to employ a filter system to ensure monochromaticity since typical Helium-Neon lasers oscillate in several axial modes. Some consideration was given to pulsed Argon II lasers because of high peak power capability and the desirable 5145A wavelength. But, as with He-Ne, the absorption cell would require excitation which is atypical of hypersonic tunnels. Further, less is known spectroscopically about highly excited Ne and ArII than CO_2 .

The experimental work can be divided into two parts: absorption and emission. The absorption experiments, which were the simpler and more successful, were undertaken to verify the results of previous researchers (Reference 11) and the theoretical predictions of the effect of a foreign gas on the center line absorption coefficient. These experiments involved a direct measurement of the absorption coefficient by a measurement of beam attenuation. They were, however, not central to a feasibility demonstration as explained above. The emission experiments were to provide this demonstration. In these experiments, the absorption coefficient was determined from a measurement of the re-emitted laser light. This phase of the work met with only limited success. The experiments were constantly plagued by background blackbody radiation and difficulties arising from the extremely long lifetime of the rotational-vibrational state of CO_2 being used. These difficulties will be discussed below.

APPARATUS

(i) Absorption Measurements

Figure 2 is a line drawing of the absorption-foreign gas broadening apparatus. This apparatus consisted of an absorption cell, laser, slug-calorimeter, and supporting equipment.

The laser was a Resalab 5 watt, CW, CO₂, water cooled laser oscillating at 10.6 μ . A piezoelectric transducer was mounted on the rear mirror of the laser to vary the cavity length and thereby the frequency.

The absorption cell consisted of a 102cm length of borosilicate glass tubing, 15.0mm in diameter. The ends of the absorption tube were closed by NaCl flats 25mm in diameter. The tube was routed at the center through a stopcock to a mechanical fore-pump and oil diffusion pump and CO₂ and He gas supplies.

After passing through the absorption cell, the laser beam was focused on a slug calorimeter using a single convex lens. This was necessary because the 10.6 μ beam diverges rather rapidly. Provided the temperature change of the calorimeter was small compared to ambient, the voltage across the output terminals was directly proportional to the power incident on it. This output voltage was read directly on a VTVM.

(ii) Emission Measurements

A line drawing of the emission apparatus is given in Figure 3. The basic apparatus consisted of a laser, emission cell, cryogenic detector, and supporting equipment.

The emission cell was 20.0mm diameter borosilicate glass tubing. One end was closed by a Wood's horn terminating in a stopcock and the other end was closed by a 2.5cm diameter NaCl flat. Two NaCl flats were mounted diametrically opposed at the center of the cell. The detector was a Philco-Ford GPC-215 consisting of a gold-doped (P-Type) germanium chip cryogenically cooled by liquid nitrogen. The detector cell had an area of 1mm x 1mm with a Barium Fluoride window.

A two window-shield arrangement was used because of the amount of blackbody radiation emitted from the cell. The lower window provided a "cold" background to the detector and the shield insured that the detector would "see" only through the lower window.

In order to obtain an AC signal, a chopper was employed. The chopper was placed between the upper window and the detector rather than between the cell and the laser. This was done due to the long lifetime (4.7 sec.) of CO₂. Had the lifetime been short

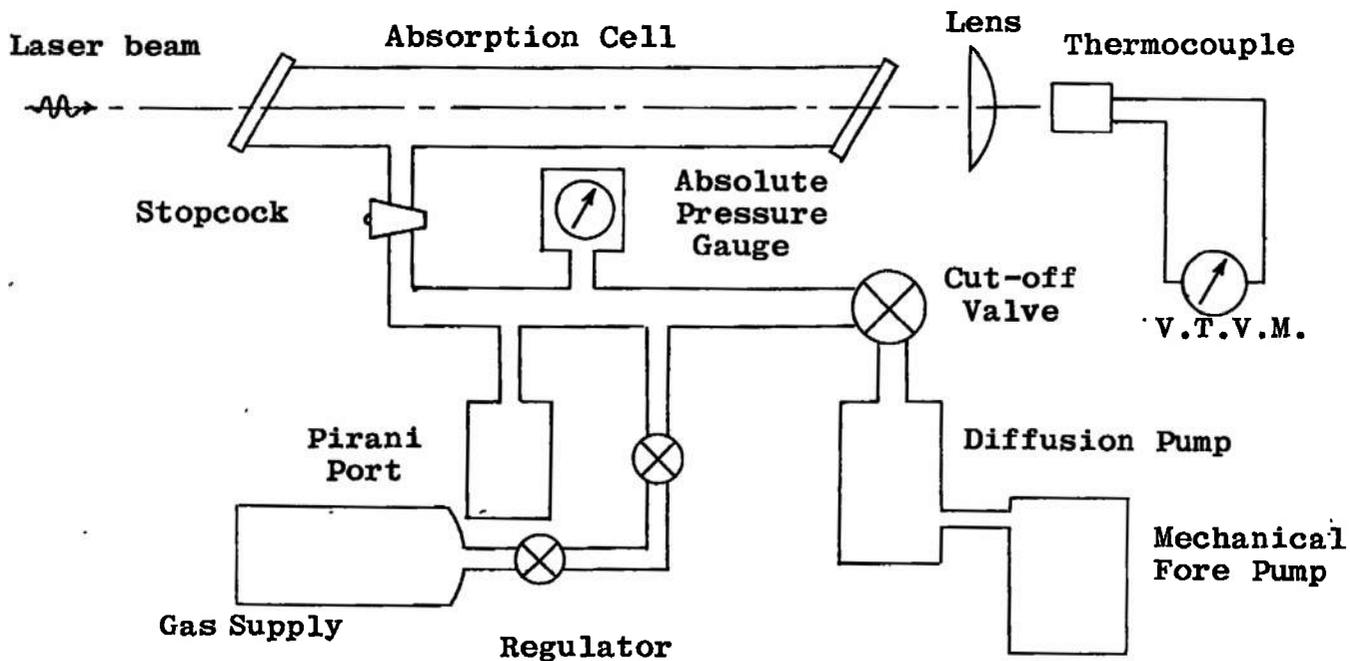


Figure 2 System diagram showing details of vacuum and instrumentation of the absorption cell.

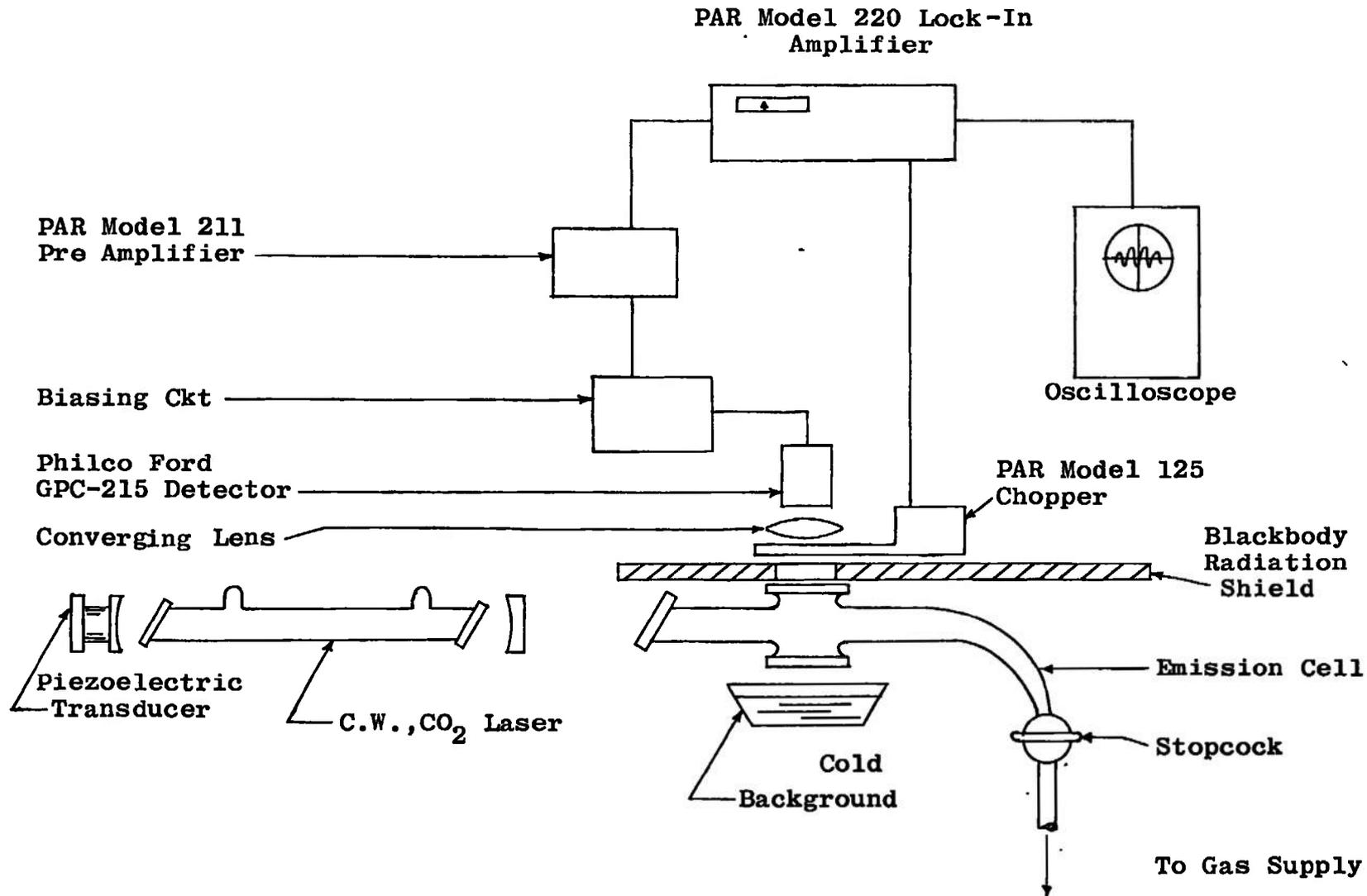


Figure 3. System diagram showing details of emission cell and instrumentation.

($\sim 10^{-8}$ sec), the chopper could have been placed between the laser and emission cell. This would have resulted in a chopped emitted light which could have been easily detected above the background. With the arrangement used, the background was also chopped making it very difficult to distinguish signal from background.

The emission cell was connected via the stopcock to an oil diffusion pump and mechanical forepump and a CO_2 gas source.

The output of the detector was fed to a differential amplifier and oscilloscope. An internal D.C. offset was provided in the differential amplifier and was used to sufficiently offset the chopped signal so that the positive pulses could be observed on the oscilloscope.

A Faraday shield was placed around the detector and associated circuitry and grounded to one side of the detector output. This arrangement greatly reduced fluctuations in the detector output.

Extraordinary care was taken to insure no ground loop currents existed.

PROCEDURE

(i) Absorption Measurements

Absorption experiments were made to check the results of previous experimenters and to validate the theoretical prediction that the pressure of foreign atoms will decrease the center line absorption coefficient.

The absorption coefficient was determined by measuring the attenuation of the laser beam over the cell when an absorbing gas is present and applying equation (5a). This attenuation was measured by measuring the output from the calorimeter with and without the absorbing gas. Assuming a linear relationship between the calorimeter output and the power incident upon it (see above), a difference in calorimeter voltages was directly proportional to the power absorbed and the calorimeter voltage without the absorbing gas was directly proportional to the incident power.

Figure 4 shows the results of absorption coefficient measurements for pure CO_2 over a range of pressures from 3 Torr to 20 Torr. Gerry and Leonard's results are given for comparison.

Figure 5 gives the absorption coefficient with a constant partial pressure of CO_2 (4 Torr) and an increasing partial pressure of He. The predicted decrease in the absorption coefficient

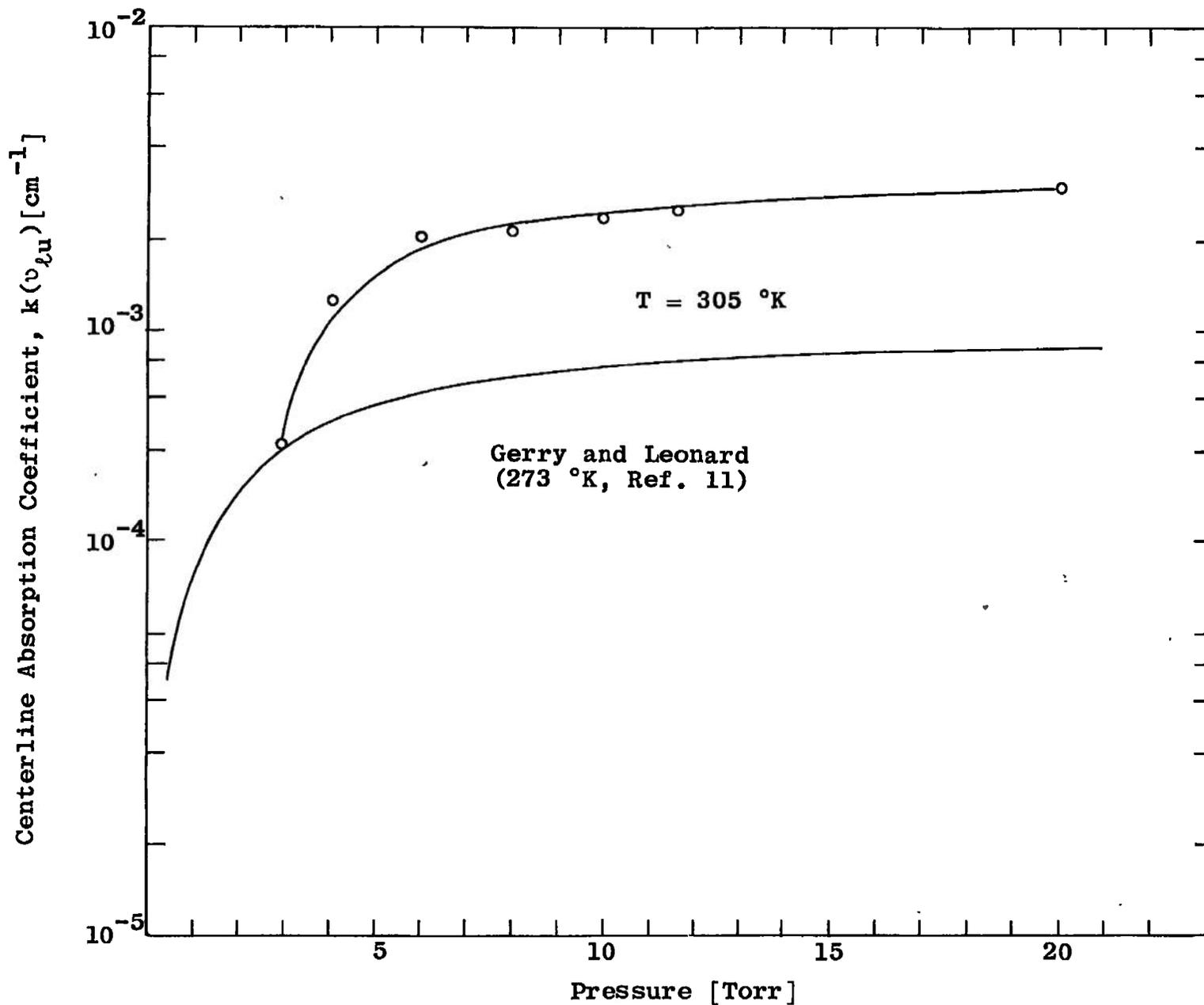


Figure 4. Centerline absorption coefficient of pure CO_2 as a function of pressure

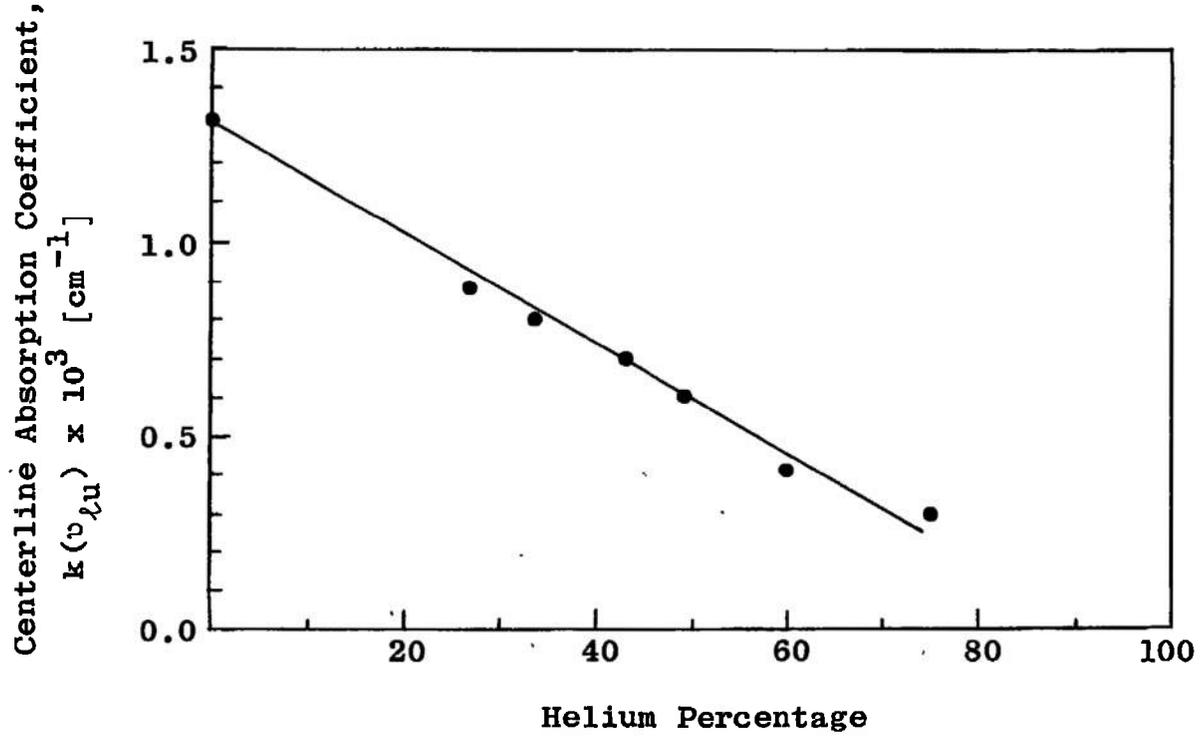


Figure 5. Centerline absorption coefficient of CO₂-He as a function of percentage He.

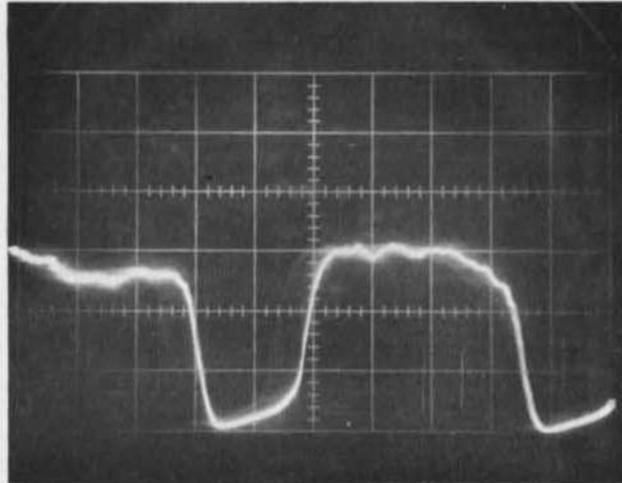
is evident.

(ii) Emission Measurements

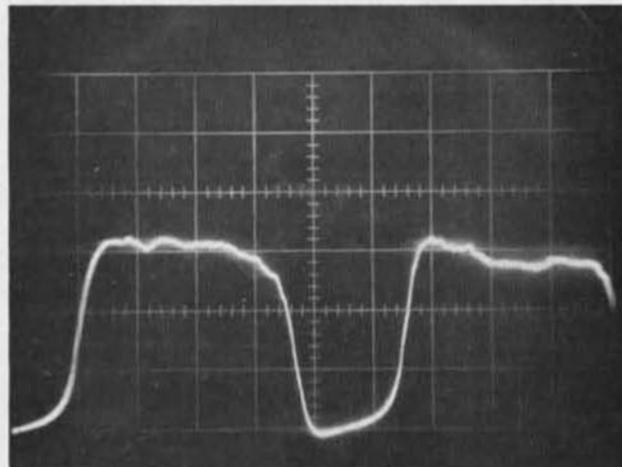
Based on preliminary calculations of emitted radiation (see Appendix A), $180\mu\text{v}$ was expected as detector output. These calculations were based on the use of a collecting lens as in Figure (2) and assumptions of detailed balancing, isotropic re-emission, and detection of all photons emitted into a solid angle, Ω , intersected by the detector. However no emitted radiation was detected. On a careful analysis of the apparatus and the physical situation, it was decided to dispense with the lens because of its relatively poor quality and questionable transmission properties. Also focusing difficulties of the lens on the small area of the detector chip were felt to compromise the experiment. Further evaluation of the long lifetime of CO_2 (4.7 sec) (Ref. 11) revealed that the lens was not focusing emitted radiation from a small volume since the entire volume in the angle, Ω , was filled with emitters. It was felt that better results could be obtained by simply placing the detector as close as possible to the emission cell.

With this new arrangement, measurements were made on emitted radiation with 20 Torr of CO_2 in the emission cell. After (on the order of) 5 seconds a maximum signal of $10\mu\text{v}$ was observed. In Figure 6, the chopped signal is shown without and with the laser on. The increase in peak excursion is attributable to reemission. Figure 7 contains the same information but with the time scale collapsed. This was considerably less than the $180\mu\text{v}$ predicted for the previous arrangement. However, an analysis based on the new geometry (Appendix A), but retaining the previous assumptions, gives a predicted output of $47.5\mu\text{v}$. Although these predictions cannot be taken too seriously since the long lifetime of the state guarantees that the emitters will not all be in the absorbing volume, but will probably fill the whole cell, they may still be used as guides. Therefore, it is believed that the $10\mu\text{v}$ signal actually represented emitted radiation.

Further experimentation was obviated because the expensive detector was damaged beyond repair toward the end of the contractual work period.

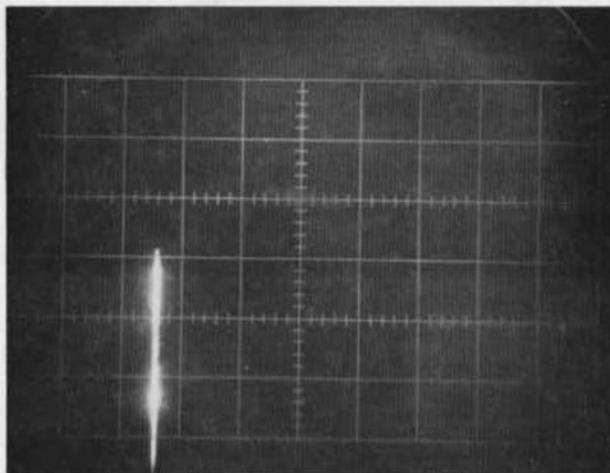


a) Detector output with laser off (blackbody background)
Vertical Scale: 50 $\mu\text{v}/\text{cm}$

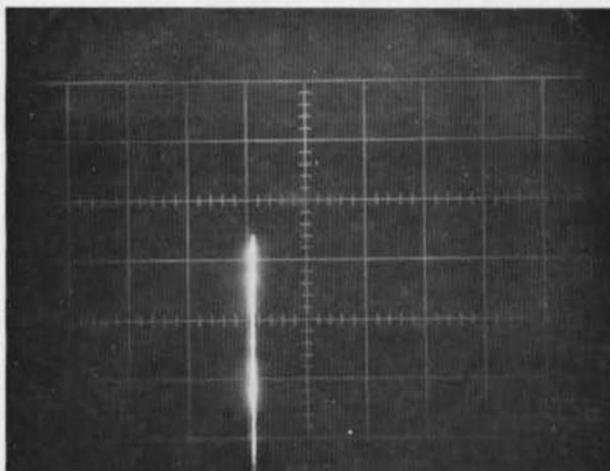


b) Detector output with laser on (blackbody plus emission)
Vertical Scale: 50 $\mu\text{v}/\text{cm}$

Figure 6 Measurements of reemitted radiation
(Time scale expanded)



a) Detector Output with laser off (blackbody background)
Vertical Scale: 50 $\mu\text{v}/\text{cm}$



b) Detector Output with laser on (blackbody plus emission)
Vertical Scale: 50 $\mu\text{v}/\text{cm}$

Figure 7 Measurements of Reemitted Radiation
(Time Scale collapsed)

SECTION IV

DISCUSSION AND CONCLUSIONS

The discussion in the preceding section has not been presented in chronological order. The emission experiments were actually attempted first and it was not until late in the project that absorption measurements were performed. In retrospect, it is apparent that the absorption measurements should have been performed before emission measurements were attempted. Had this been done, much more could possibly have been learned about the technique. For example, the line could have been swept by the laser and the basic idea could have been tested under adverse conditions such as in the presence of an electrical discharge, under various pressure conditions, and in the presence of more than one gas species. Such a course would naturally have opened up its own set of problems such as stabilization of the laser and the development of a sweeping technique. But the experiments would have been comparatively simple and the results, perhaps, of considerable practical value.

However, there were good reasons to expect success with emission experiments. Particularly, the value of the absorption coefficient for CO_2 obtained by Gerry and Leonard implied that the emitted signal was detectable (Reference 11 and Appendix A). Also, it was felt that sufficient work had been done in the absorption case to conclude that an average density measurement using absorption was feasible and hence that nothing fundamental would be gained by performing such a measurement. The real problem was to demonstrate the feasibility of density measurements using emission. Although this reasoning was sound, unforeseen problems resulting primarily from blackbody radiation and the fact that the lifetime of the state being considered was so long made the experiments exceedingly difficult.

Of these problems, the most foreboding is the blackbody radiation problem. Even under controlled laboratory conditions, this background radiation was of sufficient magnitude and fluctuated in such a fashion as to make detection of emitted coherent radiation practically impossible. These conditions will most assuredly be aggravated by a hostile environment. Therefore, the success of the technique may depend to a large degree upon the possibility of selectively filtering out this background radiation.

Detectability will undoubtedly be improved considerably if the upper state of the transition being studied has a very short lifetime ($\sim 10^{-8}$ sec). If this is so, the laser beam itself may be chopped and, since reemission occurs in an extremely short time, the emitted radiation alone (not the background) will be chopped. The problem is then reduced to detecting a square wave

of known frequency above a randomly fluctuating background. As was pointed out in the preceding section, such a procedure was not possible in the case of the 10.6μ line of CO_2 because of the long lifetime of the upper state (4.7 sec).

However, even with a short lifetime of the upper state and the subsequent ability to obtain a chopped signal, the application of this technique will be confronted with a magnification of one problem appearing in the laboratory. That being arrangement of the optical system so that a sufficient number of emitted photons are detected. In the laboratory it is possible to place the detector and collecting lens relatively close to the laser beam so that the acceptance cone (solid angle Ω , Appendix A) may be large. This may not be possible in a wind tunnel since the distance between laser beam and collecting lens is large. This may be remedied somewhat by using a system of collecting lenses and mirrors with subsequent optical complexity. However, it is felt that the final solution to this problem will have to wait for high power tunable lasers.

In the vein of increased power (and chopped input), the possibility of using pulsed lasers rather than CW lasers is brought to mind. These have been used with some success in plasma diagnostics (References 17-22). Such a laser system may provide sufficient power to make a complicated lens system unnecessary.

Any final remarks should be prefaced by the statement that the authors still feel optimistic about the ultimate success of this technique. It seems safe at this time to conclude that such a technique based on pure absorption is feasible. Based on this investigation and the fact that we have perhaps succeeded in detecting some emission, it is felt that with some more concentrated and well directed work, the emission technique can be proven feasible.

SECTION V

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APPENDIX A

The geometry of the experimental configuration using the lens is given in Figure 8. The power incident upon the detector is given by

$$P_{\text{detect}} = \frac{\Omega}{4\pi} k(\omega) P_L(\omega) dx$$

where the following assumptions were utilized:

- (1) All photons absorbed in the scattering volume reemit therein; i.e., detailed balancing with respect to absorption and emission.
- (2) Isotropic reemission
- (3) All photons emitted into Ω are detected; i.e., neglect reabsorption processes.

For the apparatus shown,

$$P_{\text{detect}} \approx 2.4 \times 10^{-3} k(\omega) P_L(\omega) dx$$

Using the centerline absorption coefficient for pure CO_2 :

$$k(\nu_{\ell u}) = 1.5 \times 10^{-4} \text{cm}^{-1}$$

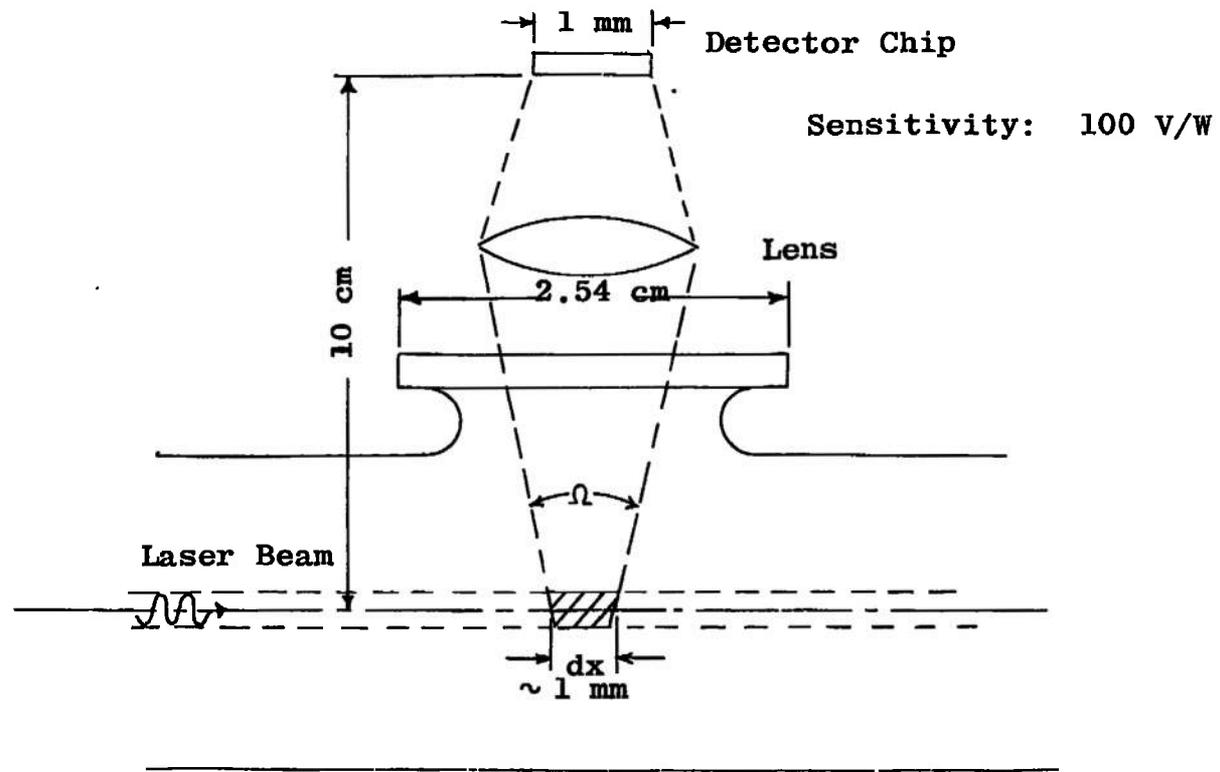
and a laser power

$$P_L = 5 \text{ W},$$

we have for the power incident on the detector chip

$$P_{\text{detect}} = 1.8 \mu\text{W}$$

and $V_{\text{signal}} = 180 \mu\text{V}$



a) Experimental Configuration

Figure 8.

Figure 9 gives the second arrangement of the emission cell and detector. Here a portion of the radiation absorbed over the length x_0x_1 will be detected. To calculate the amount of detected radiation, proceed as follows:

From the geometry

$$e = \frac{(b-l)(d+c)}{2c}$$

Assuming $\alpha \approx \beta$, the detected power from a small element, dx , of x_0x_1 is

$$P_{\text{detect}} = \frac{\beta}{4\pi} k P_0 dx$$

where

P_0 = incident laser power

k = absorption coefficient

$$\beta \approx \frac{l^2}{(c+d)^2}$$

Then

$$\begin{aligned} P_{\text{detect}} &\approx \frac{l^2}{4\pi(c+d)^2} k P_0 \int_{-e-\frac{l}{2}}^{e+\frac{l}{2}} dx \\ &\approx \frac{l^2}{4\pi(c+d)^2} k P_0 (2e+l) \end{aligned}$$

Now

$b = 2\text{cm}$ (window diameter)

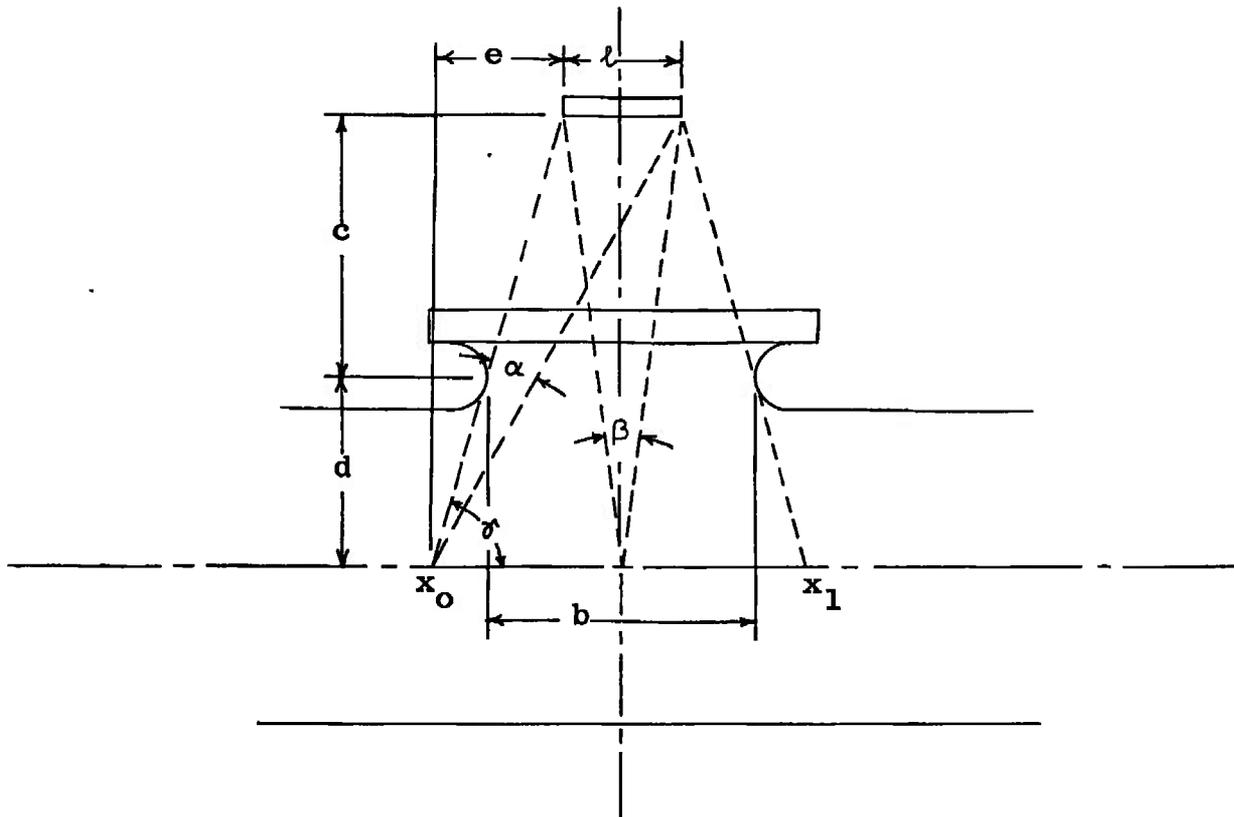
$l = 1\text{mm}$ (detector chip dimension)

$c = 1.27\text{cm}$ (distance from chip to window)

$d = 1\text{cm}$ (distance from centerline to window)

$k = 1.5 \times 10^{-4} \text{cm}^{-1}$ (absorption coefficient)

$P_0 = 5 \text{ watts}$



b) Experimental Configuration
Figure 9.

Then

$$P_{\text{detect}} = 0.475 \text{ } \mu\text{W}$$

and

$$V_{\text{signal}} = 47.5 \text{ } \mu\text{V}$$

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13. ABSTRACT A feasibility study of a technique to measure gas densities using resonant scattering of laser light is described. The technique involves the illumination of the gas by a laser beam whose frequency is matched to a natural transition frequency of one species of the gas. The re-emitted light is then detected. The density is obtained from the integrated absorption coefficient which is determined by sweeping the laser frequency over the line. A theoretical description is given of line broadening mechanisms. Experimental studies were initiated involving both absorption and reemission of coherent radiation.			

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	ROLE	WT	ROLE	WT	ROLE	WT
resonant scattering resonant absorption laser gas density						

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