NASA CR-132769

UCLA-ENG-7338
May 1973

N73-28554

THE APPLICATION OF FAR I.R. LASERS TO THE INVESTIGATION OF THE OPTICAL PROPERTIES OF MATERIALS

O. M. STAFSUDD

CASE FILE
COPY

FINAL REPORT

NASA GRANT N.G.L. – 05-007-161

School of Engineering and Applied Science
University of California
Los Angeles, California

PREFACE

During the final period of this grant a theoretical and experimental study was made of the effect of diffusion on the saturation of the infrared absorption of radiation by gases. This report contains most of the theoretical development of a model of a gas absorber including diffusion and a small amount of the experimental evidence we gathered to support this model.

The experimental results along with the theoretical model will be submitted for publication in the near future.

Abstract

Effect of diffusion on the saturation and absorption of the molecular gases is considered. The analysis starts with a simple derivation of the multi-level rate equations. Incorporating the diffusion term, calculations are carried out both for the homogeneous and inhomogeneous broadening cases. Both graphical and analytical solutions are obtained for the absorption coefficient and the saturation intensity. It is shown that the effect of diffusion is characterized by a diffusion function $K'(k_2a)$, which varies inversely with the radius of the beam at low pressures and small beam sizes. The analysis is done in terms of two parameters k_1a and k_2a, which are simple functions of the intensity of the beam I, the effective relaxation rate w, and the effective diffusion constant D. "a" is the radius of the beam.

I. Introduction

Absorption and saturation of infrared radiation around 10.6μ in molecular gases has recently been of considerable interest.[1,2,3,4] Using a two-level model,[5] where one is only concerned with the lower and upper levels used in absorption, the variation of the laser beam intensity, I, is found to be

$$\frac{dI}{dz} = -\alpha I \qquad (1)$$

where α is the absorption coefficient which can be expressed as

$$\alpha = \frac{\alpha_0}{\left[1 + \frac{I}{I_s}\right]^n} \qquad (2)$$

In this expression I_s is the saturation intensity; α_0 is the small signal absorption coefficient; n is 1 when the transition is homogeneously broadened and .5 when the transition is inhomogeneously broadened.

If we define

$$\begin{aligned} M &= M_1 + M_2 \\ \Delta M &= M_2 - M_1 \qquad (3) \\ m &= \Delta M/M \end{aligned}$$

where M_1 and M_2 are the lower and upper level populations, equation (2) can be written as[6]

$$m = -\frac{1}{1 + \frac{I}{I_s}} \tag{4}$$

Even though two-level model gives a very simple explanation of saturation, it is not totally satisfactory because of various phenomena that are involved in saturable gases. The most important effects are the tight coupling between various vibrational-rotational levels and the spatial diffusion of molecules. If multirotational levels are considered, C.P. Christensen et al[7] have showed that the results of the two-level model can be used by replacing relaxation rates and diffusion constants by their effective values as follows:

$$\begin{aligned} w_2 &= \frac{\sum_i w_i^u \, m_i^u}{M_2} \\ w_1 &= \frac{\sum_i w_i^e \, m_i^l}{M_1} \\ D_2 &= D \, \frac{\sum_i m_i^u}{M_2} \\ D_1 &= D \, \frac{\sum_i m_i^l}{M_1} \end{aligned} \tag{5}$$

In these expressions w_1 and w_2 are the lower and upper effective relaxation rates; $w_i^{u,l}$ is the ith rotational level relaxation rate in the upper(lower) band;

$m_i^{u,l}$ is the corresponding population density; M_1 and M_2 are the lower and upper absorption level population densities. Similarly, D_1 and D_2 are the effective diffusion constants, and D is the diffusion constant. Assuming the rotational constant B for the upper and lower bands are the same, D_1 and D_2 will be nearly equal. A more simple derivation of these expressions than was done originally is included in the appendix.

With the diffusion term included, the rate equation for the inversion density can be obtained from equation (4) as[6]

$$D\nabla^2 m - (2\sigma I + w)m = w \qquad (6)$$

where D is assumed to be the effective relaxation rate for both bands; w is equal to w_2; σ is the absorption cross-section at the laser wavelength, and I is the intensity of the beam in photons/cm^2-sec. Since the lower level for most saturable absorbers is the ground state, w_1 is assumed to be zero.

The absorption coefficient can be written as

$$\alpha = -\frac{1}{I}\frac{dI}{dz} = m\alpha_o \qquad (7)$$

where α_o is the small signal absorption coefficient.

Since all the variables are a function of the radial coordinate r, we would like to define average quantities. First of all, average intensity I_{av} is defined as

$$P = \pi a^2 I_{av} \tag{8}$$

where P is the power and a is the radius of the beam. Then equation (7) can be transformed to

$$\alpha_{av} = - \frac{1}{I_{av}} \frac{dI_{av}}{dz} = \alpha_o m_{av} \tag{9}$$

where

$$m_{av} = \frac{\int_0^\infty m e^{-r^2/a^2} 2\pi r dr}{a^2} \tag{10}$$

for a Gaussian beam. If the beam is rectangular instead of Gaussian as discussed in the next section, the equations would be the same except for

$$m_{av} = \frac{\int_0^a m\ 2\pi\ r dr}{a^2} \tag{11}$$

II. Rectangular Wave Approximation

In order to simplify the analysis, we will assume a rectangular intensity distribution in the form

$$I = \begin{cases} I & \text{for } 0 \leqq r \leqq a \\ 0 & \text{for } r > a \end{cases} \qquad (12)$$

Then the differential equation becomes

$$\frac{d^2}{dr^2} m(r) + \frac{1}{r} \frac{d}{dr} m(r) - \frac{(2\sigma I + w)}{D} m(r) = \frac{w}{D} \qquad o \leqq r \leqq a$$

$$\frac{d^2}{dr^2} m(r) + \frac{1}{r} \frac{d}{dr} m(r) - \frac{w}{D} m(r) = \frac{w}{D} \qquad r > a \qquad (13)$$

Letting

$$k_1^{\,2} = \frac{2\sigma I + w}{D}$$

$$k_2^{\,2} = \frac{w}{D} \qquad (14)$$

We obtain the solution

$$m = \begin{cases} m_1 & o \leqq r \leqq a \\ m_2 & r > a \end{cases} \qquad (15)$$

$$m_1 = AI_o(k_1 r) + BK_o(k_1 r) - \frac{k_2^{\,2}}{k_1^{\,2}}$$

$$m_2 = CI_o(k_2 r) + K_o(k_2 r) - 1$$

$I_0(x)$ is the modified Bessel function of the first kind of zero order and $K_0(x)$ is the modified Bessel function of the second kind of zero order. A, B, C, and D are constants to be determined by boundary conditions, which can be taken as

$$\begin{aligned} &(1)\quad m_1(a) = m_2(a) \\ &(2)\quad \dot{m}_1(a) = \dot{m}_2(a) \\ &(3)\quad m_1(0) = 0 \\ &(4)\quad m_2(b) = -1 \end{aligned} \qquad (16)$$

where b is the radius of the cylindrical absorption cell.

Boundary condition (3) dictates B to be zero. Using the other boundary conditions, and equation (11), we obtain[6]

$$m_{av} = \frac{2\dfrac{(k_2^2 - k_1^2)}{k_1^2} \dfrac{I_1(k_1 a)}{k_1 a}}{I_0(k_1 a) + \dfrac{F}{E} k_1 I_1(k_1 a)} - \frac{k_2^2}{k_1^2} \qquad (17)$$

where I(x) is the first order modified Bessel function of the first kind and

$$\begin{aligned} E &= k_2 \left[I_1(k_2 a) K_0(k_2 b) + K_1(k_2 a) I_0(k_2 b) \right] \\ F &= \left[K_0(k_2 a) I_0(k_2 b) - I_0(k_2 a) K_0(k_2 b) \right] \end{aligned} \qquad (18)$$

By definition, the saturation intensity occurs when $m_{av} = -\frac{1}{2}$. Then we find

$$\frac{k_1{}^2 - k_2{}^2}{k_1 a(k_1{}^2 - 2k_2{}^2)} = \frac{1}{4}\left[\frac{I_o(k_1 a)}{I_1(k_1 a)} + \frac{F}{E}k_1\right] \qquad (19)$$

It can be shown that the terms with $K_o(k_2b)$ are negligible for reasonable values of b. For example, in case of SF_6, k_2 is about 1.17 at 1 Torr.[8] If we take b to be 1 inch, then $k_2b = 2.98$. However, k_2 is essentially proportional to the pressure, as will be shown later, so that k_2b will be smaller at smaller pressures. Let us take two extreme cases:

<u>Case 1.</u> $k_2b = .1$ $\quad$ $k_2a = .01$

$K_o(k_2b) = 2.427$ $\quad$ $K_1(k_2a) = 100$

$I_1(k_2a) = 0$ $\quad$ $I_o(k_2b) = 1$

$K_o(k_2a) = 50$ $\quad$ $I_o(k_2a) = 1$

Therefore $I_1(k_2a)\ K_o(k_2b) << K_1(k_2a)\ I_o(k_2b)$ and $K_o(k_2a)\ I_o(k_2b) >> I_o(k_2a)\ K_o(k_2b)$

<u>Case 2.</u> $k_2b = 4$ $\quad$ $k_2a = .4$

$K_o(k_2b) = .011$ $\quad$ $K_1(k_2a) = 2.18$

$I_1(k_2a) = .2$ $\quad$ $I_o(k_2b) = 11.3$

$K_o(k_2a) = 1.115$ $\quad$ $I_o(k_2a) = 1.04$

Again $I_1(k_2a)\ K_o(k_2b) << K_1(k_2a)\ I_o(k_2b)$ and $K_o(k_2a)\ I_o(k_2b) >> I_o(k_2a)\ K_o(k_2b)$

So ignoring $K_o(k_2b)$, we obtain

$$E \overset{o}{=} I_o(k_2b)\ K_1(k_2a)\ k_2$$

$$F \overset{o}{=} I_o(k_2b)\ K_o(k_2a) \qquad (20)$$

Then equation (19) can be written as

$$\frac{k_1^2 - k_2^2}{k_1 a\,(k_1^2 - 2k_2^2)} = \frac{1}{4}\left[\frac{I_o(k_1a)}{I_1(k_1a)} + \frac{K_o(k_2a)}{K_1(k_2a)}\,\frac{k_1}{k_2}\right] \qquad (21)$$

Let us call the right-hand side of this equation Z. If we let

$$X = \frac{(k_1a)^2}{(k_2a)^2} = \frac{k_1^2}{k_2^2} \qquad (22)$$

We have

$$X = \frac{1 - 2(k_1a)Z}{1 - (k_1a)Z} \qquad (23)$$

If we plot equations (22) and (23) together, as a function of (k_1a) for various values of (k_2a), the intersection point gives the value of k_1a as well as X. Then the saturation intensity is

$$I_s = \frac{w}{2\sigma}\left[X - 1\right] h\nu \qquad (24)$$

Figure 1 is the result of these calculations. The intersection points determine X as a function of k_2a. $(X - 1)$ versus k_2a is plotted in Figure 2. For a parti-

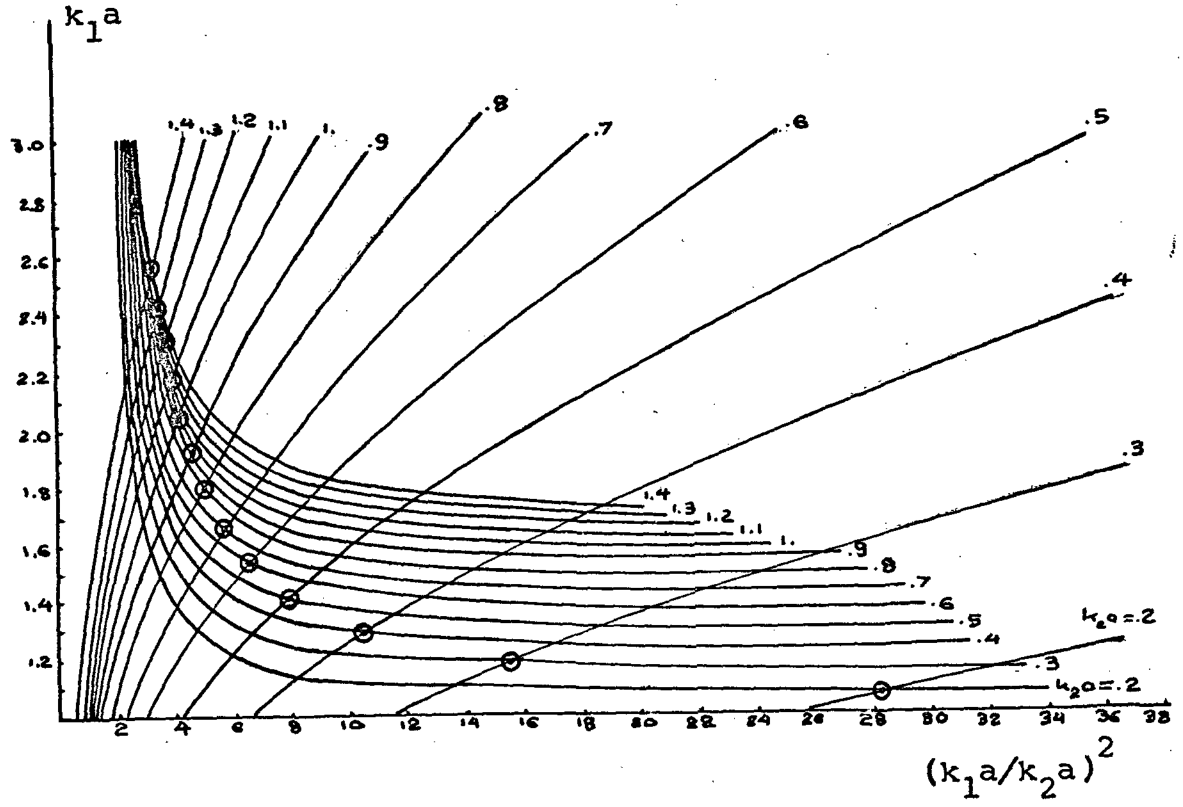

FIGURE 1

GRAPHICAL METHOD FOR THE CALCULATION

OF THE SATURATION INTENSITY

cular gas k_2 is equal to $\sqrt{w/D}$. Figure 2 shows that the saturation intensity changes very drastically for small values of k_2a. For large values of k_2a, the saturation intensity does not change much.

We note that the theory includes the effect of the cylindrical absorption cell walls with the boundary condition $m(b) = -1$.

III. Analytical Derivation of the Absorption Coefficient and the Saturation Intensity

Experimentally we normally measure the output power as a function of the input power, and deduce the variation of absorption as the intensity of the beam is varied. So an important quantity to know is m_{av} as a function of the input power. We can write equation (17) as[6]

$$m_{av} = \frac{(1 - 2k_1 a\, Z) - X}{(2k_1 a\, Z)\, X} \tag{25}$$

m_{av} as a function of $(X - 1)$ is plotted in Figure 3 for various values of $k_2 a$. We see that the absorption decreases much faster as the beam size is increased.

Without diffusion, m_{av} changes as

$$m_{av} = -\frac{1}{1 + I/I_s} \tag{26}$$

Differentiating this expression with respect to I we obtain

$$\overset{o}{m}_{av} = \frac{1}{I_s}\,\frac{1}{(1 + \frac{I}{I_s})^2} \tag{27}$$

At I = 0, we have

$$\overset{o}{m}_{av}\,(o)\, I_s = 1 \tag{28}$$

When diffusion is included, we should obtain the same result as I goes to zero. Now we have[6]

$$\overset{o}{m}_{av}(o) = \frac{2\sigma}{w}\left[1 - \frac{2}{k_2a\left[\frac{I_o(k_2a)}{I_1(k_2a)} + \frac{F}{E}\right]}\right] \qquad (29)$$

Approximating F/E as before and comparing equations (28) and (29) we obtain

$$I_s = \frac{w}{2\sigma} \; K(k_2a) \qquad (30)$$

where the function $K(k_2a)$ is given by

$$K(k_2a) = \frac{1}{1 - \frac{2}{k_2a\left(\frac{I_o(k_2a)}{I_1(k_2a)} + \frac{K_0(k_2a)}{K_1(k_2a}\right)}} \qquad (31)$$

$w/2\sigma$ is the multi-level saturation intensity. The function $K(k_2a)$ is what we called $(X - 1)$ before. It is the factor that is caused by the diffusion and wall effects, and is plotted in Figure 2. It is seen that the graphical and analytical solutions give essentially the same results.

Experimentally we normally measure output power versus input power. It is easier to do this measurement at high power densities. For example, if $I_{measured}$ =

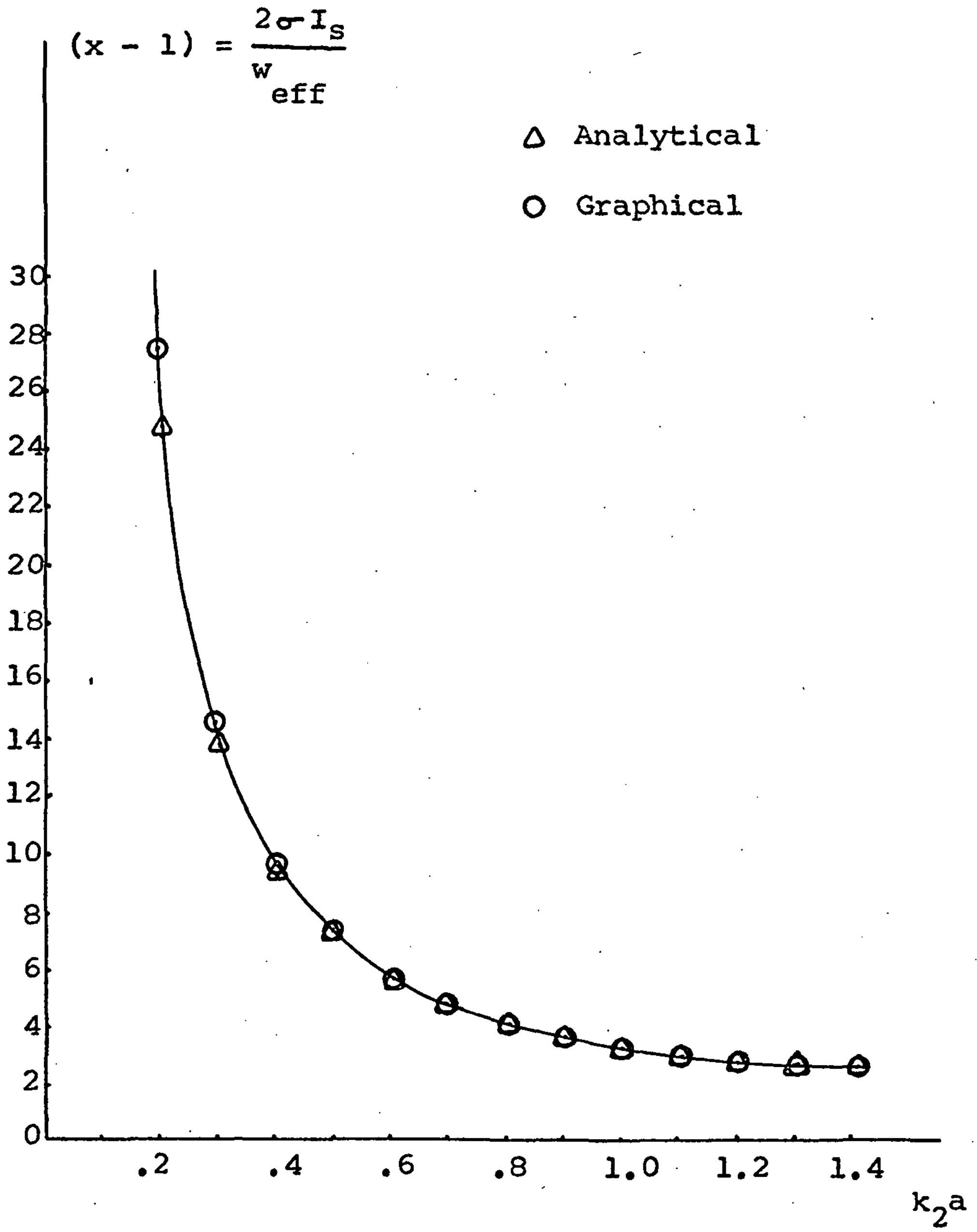

FIGURE 2

GRAPHICAL AND ANALYTICAL SOLUTIONS

FOR THE SATURATION INTENSITY OF MOLECULAR GASES

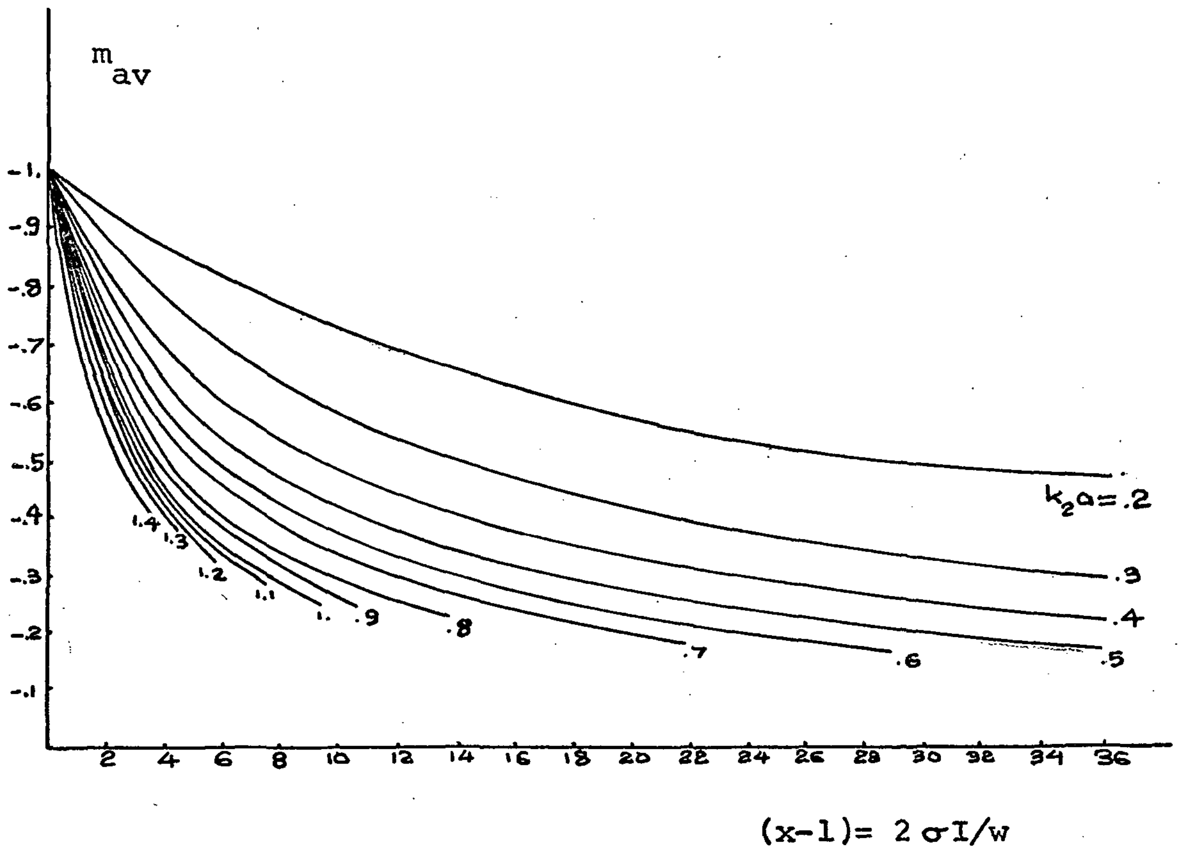

FIGURE 3

INVERSION DENSITY AS A FUNCTION OF INTENSITY

2.1 W/cm^2 and I_{real} = 2.2 W/cm^2, we have % error = 5%. At low power densities, if $I_{measured}$ = .0001 W/cm^2 and I_{real} = .0002 W/cm^2, % error = 50%. However, we need to show how to interpret the results at high power densities meaningfully. At large intensities, we can write

$$k_1{}^2 \cong \frac{2\sigma I}{D} \tag{32}$$

This is further justified in the next section. Now we can write equation (25) in the form[6]

$$m_{av} = - \frac{\left[\frac{D}{W}\,\frac{1}{Z'} + 1\right]}{\frac{2\sigma I}{W} + 1} \tag{33}$$

where

$$Z' = \frac{2a}{k_1}\, Z \tag{34}$$

For large values of k_1, we can write

$$Z' = \frac{1}{2}\,\frac{K_o(k_2 a)}{K_1(k_2 a)}\,\frac{a}{k_2} \tag{35}$$

This is further justified in the next section. Combining equations (9), (32), and (35), we obtain

$$\frac{1}{I}\,\frac{dI}{dz} = - \frac{K'(k_2 a)}{\frac{2\sigma I}{W} + 1}\,\alpha_o \tag{36}$$

where

$$K'(k_2a) = \frac{2K_1(k_2a)}{k_2a\,K_o(k_2a)} + 1 \tag{37}$$

Integrating equation (36), we find

$$K'(k_2a)\,\alpha_o L = \ln \frac{I_{out}}{I_{in}} + \frac{2\sigma}{w}\left[I_{out} - I_{in} \right] \tag{38}$$

At high power densities, the first term on the right-hand side can be neglected. Then we have

$$I_{out} = I_{in} - \frac{w}{2\sigma}\,K'(k_2a)\,\alpha_o L \tag{39}$$

Without diffusion, we would have

$$\frac{1}{I}\,\frac{dI}{dz} = -\,\frac{\alpha_o}{1 + \frac{I}{I_s'}} \tag{40}$$

Neglecting 1 in the denominator, we would obtain

$$I_{out} = I_{in} - \alpha_o L\, I_s' \tag{41}$$

Therefore, it is tempting to identify

$$I_s' = \frac{w}{2\sigma}\,K'(k_2a) \tag{42}$$

The actual saturation intensity is given by equation (30). Therefore if one measures I_s' experimentally, the actual saturation intensity will be

$$I_s = [CF]\ I_s' \tag{43}$$

where [CF] is the correction factor. It is given by

$$[CF] = \frac{K(k_2a)}{K'(k_2a)} \tag{44}$$

[CF], $K(k_2a)$, and $K'(k_2a)$ are tabulated in Table 1.

TABLE 1

Functions of Interest

k_2a	$x - 1$	$\frac{2K_1(k_2a)}{K_0(k_2a)k_2a} + 1$	CF
0.1	75.075	82.202	0.9133
0.2	24.39	28.248	0.8634
0.3	13.74	15.845	0.8672
0.4	9.191	10.8001	0.851
0.5	6.865	8.1672	0.841
0.6	5.473	6.5835	0.831
0.7	4.571	5.543	0.825
0.8	3.935	4.811	0.818
0.9	3.481	4.278	0.814
1.0	3.132	3.865	0.81
1.1	2.836	3.5323	0.803
1.2	2.639	3.274	0.806
1.3	2.454	3.062	0.801
1.4	2.325	2.887	0.805
1.5	2.195	2.729	0.804
1.6	2.092	2.601	0.804
1.7	2.004	2.489	0.805
1.8	1.928	2.392	0.806
1.9	1.86	2.306	0.807
2.0	1.802	2.229	0.808
2.1	1.751	2.16	0.811
2.2	1.705	2.103	0.811
2.3	1.663	2.045	0.813
2.4	1.626	1.986	0.815
2.5	1.592	1.949	0.817
2.6	1.562	1.907	0.819
2.7	1.533	1.867	0.821
2.8	1.508	1.83	0.824
2.9	1.484	1.795	0.827
3.0	1.465	1.772	0.827
3.1	1.444	1.741	0.829
3.2	1.428	1.716	0.832
3.3	1.411	1.693	0.833
3.4	1.395	1.668	0.836
3.5	1.38	1.647	0.838
3.6	1.367	1.628	0.84
3.7	1.355	1.61	0.842
3.8	1.343	1.59	0.845
3.9	1.332	1.574	0.846
4.0	1.321	1.558	0.848

IV. The Case of Inhomogeneous Broadening

At low pressures that are used with the saturable absorbers, we would expect the Doppler broadening to be dominant over collisional broadening. In order to see what happens in this case, let us rewrite equation (36) in the form

$$= \frac{L(k_1 a, k_2 a)}{\frac{2\sigma I}{w} + 1} \alpha_o \tag{45}$$

where

$$L(k_1 a, k_2 a) = \frac{2}{k_2 a} \frac{1}{\frac{I_o(k_1 a)k_2}{I_1(k_1 a)k_1} + \frac{K_o(k_2 a)}{K_1(k_2 a)}} + 1 \tag{46}$$

We need to integrate equation (45) in order to average over the classes of molecules that are broadened differently. The variables are α_o, σ, and k_1. The integral is

$$\alpha_i = \int_{-\infty}^{\infty} \alpha_h(\nu_o) p(\nu_o) dr_o \tag{47}$$

The letters h and i stand for homogeneous and inhomogeneous respectively. If the function $L(k_1 a, k_2 a)$ were independent of ν_o, then the above integral would integrate as in the classical case to give a result similar to equation (2). So let us see how this function behaves. If the beam intensity is 1 W/cm^2, we would have for SF_6,

$$2\sigma I = 4.07 \cdot 10^5$$

at the peak of the Lorentzian $g(\nu_0)$. The relaxation rate of SF_6 at 1 mm is about 50/sec[8]. However, we need to know the effective relaxation rate. It is clear, though, that $2\sigma I$ is much larger than w. So at intensities larger than the saturation intensity, it is safe to say that

$$k_1 \doteq \sqrt{\frac{2\sigma I}{D}} \qquad (48)$$

$I_0(k_1 a)/I_1(k_1 a)$ remains close to 1 for reasonable values of $k_1 a$: $K_0(k_2 a)/K_1(k_2 a)$ is close to .5 and does not change much with $k_2 a$. We have

$$\frac{k_2}{k_1} \doteq \sqrt{\frac{w}{2\sigma I}} \qquad (49)$$

Letting

$$\sigma = C\, g(\nu) \qquad (50)$$

where C is a constant, and substituting the Lorentzian lineshape function $g(\nu)$[9] in equation (48), we find

$$\frac{k_2}{k_1} \doteq \sqrt{\frac{\pi w \left[\left(\frac{\Delta\nu}{2}\right)^2 + (\nu - \nu_0)^2 \right]}{C I \Delta\nu}} \qquad (51)$$

$g(\nu)$ falls quite rapidly from its maximum value of $2/\pi\Delta\nu$. It is less than one tenth of its maximum value when $(\nu - \nu_0)$

is equal to $\Delta\nu$. With the two level model without diffusion we would have

$$\alpha_h = - \frac{MCh\nu}{\frac{1}{g(\nu)} + \frac{\pi\Delta\nu}{2}\frac{I}{I_s}} \tag{52}$$

where M is the molecular density. Substituting in $g(\nu)$, we find

$$\alpha_h = - \frac{M\,Ch\nu\;\Delta\nu/2\pi}{\left[(\nu-\nu_o)^2 + \left(\frac{\Delta\nu}{2}\right)^2\right] + \frac{\Delta\nu^2}{4}\frac{I}{I_s}} \tag{53}$$

This is broader than the lineshape function $g(\nu)$ by $\sqrt{1 + I/I_s}$. Assuming I/I_s to be 100, the width of α_h would be about 10 $\Delta\nu$. We see that as I increases, the width increases while the peak value decreases.

Let us assume $k_2a = .1$, $I_o(k_1a)/I_1(k_1a) = 1$, $K_o(k_2a)/K_1(k_2a) = .5$, $w = 10^2$, $2\sigma I = 4 \times 10^6$. Then

$$\sqrt{\frac{w}{2\sigma I}} = 1/200$$

Using these values, we define

$$Y(\nu) = \frac{20}{.01\sqrt{.25 + \frac{(\nu-\nu_o)^2}{\Delta\nu^2}} + .5} + 1 \tag{54}$$

We see that the denominator above changes very little for $(r - r_o)^2/\Delta r^2 \angle 10$. This function is plotted in Figure 4. It is seen to be varying very slowly. Therefore, for all practical purposes, it is safe to assume that $L(k_1a, k_2a)$ can be assumed to be constant while integrating α_h to find α_i. Then we immediately obtain,

$$\alpha_i = \frac{L(k_1a,\ k_2a)\,\alpha_o}{\sqrt{1 + 2\sigma I/w}} \tag{55}$$

At intensities larger than $w/2\sigma$, we can neglect the modified Bessel functions of the first kind, obtaining

$$\alpha_i = \frac{K'\,(k_2a)\,\alpha_o}{\sqrt{1 + 2\sigma I/w}} \tag{56}$$

Integrating equation (56) gives[6]

$$K'(k_2a)\alpha_oL = \left[\mathrm{In}\left[\frac{(1+\frac{2\sigma I}{w})^{\frac{1}{2}} - 1}{1+\frac{2\sigma I}{w})^{\frac{1}{2}} + 1}\right] + 2(1+\frac{2\sigma I}{w})^{\frac{1}{2}}\right]_{I_{in}}^{I_{out}} \tag{57}$$

When $2\sigma I/w \gg 1$, namely at high intensities, we obtain

$$K'(k_2a)\,\alpha_oL = 2\sqrt{\frac{2\sigma}{w}}\left[\sqrt{I_{out}} - \sqrt{I_{in}}\right] \tag{58}$$

In the case of the multi-level model without diffusion we have

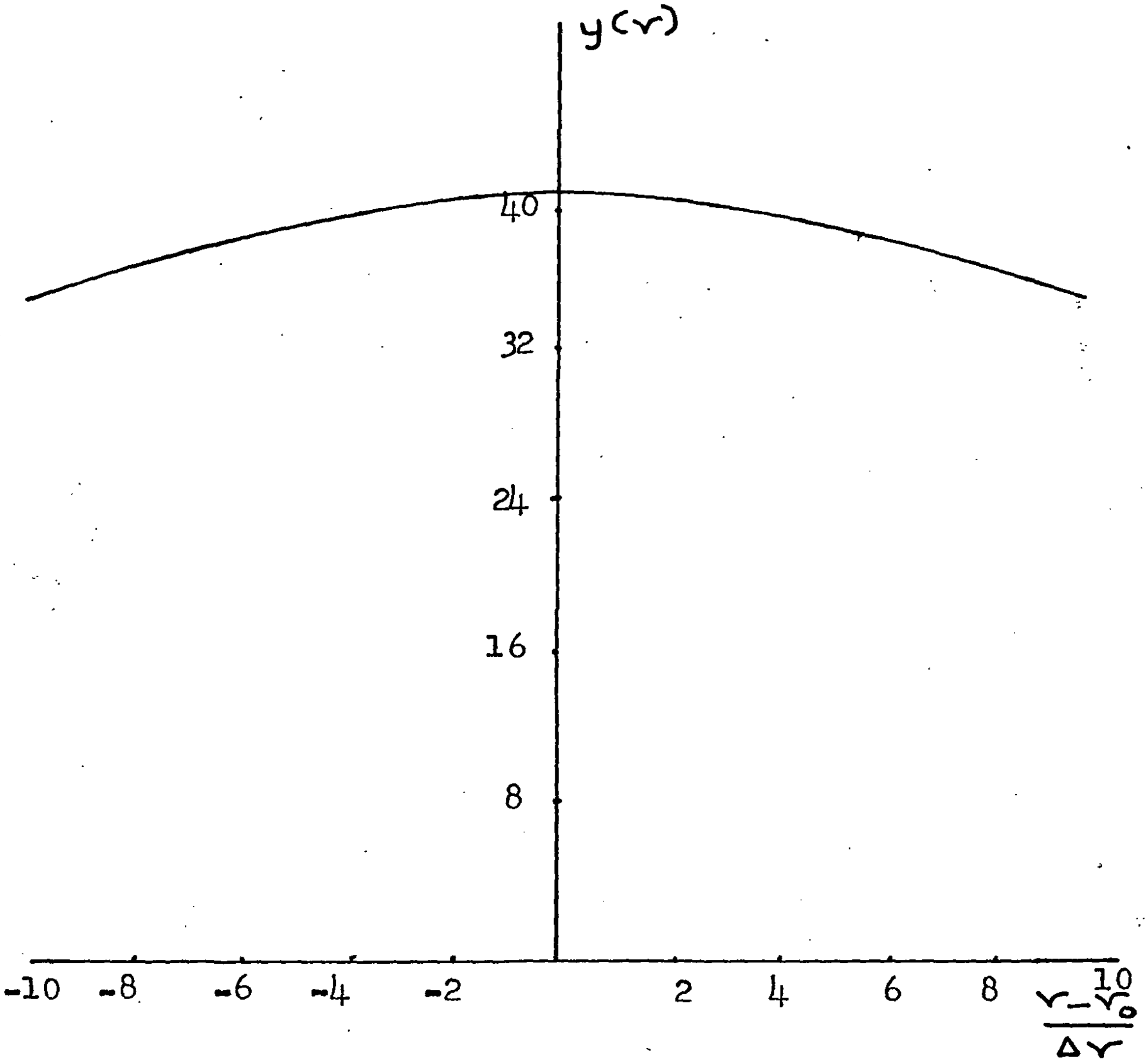

FIGURE 4

THE FUNCTION Y (r)

$$\frac{1}{I}\frac{dI}{dz} = - \frac{\alpha_0}{\left(1 + \frac{I}{I'_s}\right)^{\frac{1}{2}}} \qquad (59)$$

At high intensities, we would obtain

$$\alpha_0 L = \frac{2}{\sqrt{I'_s}} \left[\sqrt{I_{out}} - \sqrt{I_{in}} \right] \qquad (60)$$

Therefore, the saturation intensity I'_s can be defined as

$$I'_s = \left[K'(k_2 a) \right]^2 \frac{W}{2\sigma} \qquad (61)$$

The actual saturation intensity is expected to be somewhat lower than I'_s as in the homogeneous case.

V. Summary

The main results of the foregoing analysis can be summarized by the following equations:

$$k_2 = \sqrt{w/D} \tag{62}$$

w = effective relaxation rate

D = effective diffusion constant

$$k_1 = \sqrt{\frac{2\sigma I + w}{D}} \tag{63}$$

σ = absorption cross-section

I = photon intensity

a = radius of the beam

$$K(k_2a) = \frac{1}{1 - \dfrac{2}{k_2a\left[\dfrac{I_o(k_2a)}{I_1(k_2a)} + \dfrac{K_o(k_2a)}{K_1(k_2a)}\right]}} \tag{64}$$

$$K'(k_2a) = \frac{2\,K_1\,(k_2a)}{k_2a\,K_o(k_2a)} + 1 \tag{65}$$

$$L(k_1a, k_2a) = \frac{2}{k_2a} \cdot \frac{1}{\dfrac{I_o(k_1a)}{I_1(k_1a)} \cdot \dfrac{k_2a}{k_1a} + \dfrac{K_o(k_2a)}{K_1(k_2a)}} + 1 \tag{66}$$

Homogeneous Case

$$\alpha = \frac{L(k_1 a,\ k_2 a)}{\frac{2\sigma I}{w} + 1}\,\alpha_o \qquad (67)$$

$$\alpha = \frac{K'(k_2 a)}{\frac{2\sigma I}{w} + 1}\,\alpha_o \qquad I > \frac{w}{2\sigma} \qquad (68)$$

$$I_{out} - I_{in} = -I'_s\,\alpha_o\,L \qquad I > w/2\sigma \qquad (69)$$

$$I'_s = \frac{w}{2\sigma}\,K'(k_2 a) \qquad (70)$$

$$I_s = [CF]\,I'_s \qquad (71)$$

$$[CF] = K(k_2 a) / K'(k_2 a) \qquad (72)$$

Inhomogeneous Case

$$\alpha = \frac{L(k_1 a,\ k_2 a)}{\sqrt{1 + 2\sigma I/w}}\,\alpha_o \qquad (73)$$

$$\alpha = \frac{K'(k_2 a)\,\alpha_o}{\sqrt{1 + 2\sigma I/w}} \qquad I > \frac{w}{2\sigma} \qquad (74)$$

$$\sqrt{I_{out}} - \sqrt{I_{in}} = -\sqrt{I'_s}\,\alpha_o\,L/2 \qquad I > \frac{w}{2\sigma} \qquad (75)$$

$$I'_s = \frac{w}{2\sigma}\left[K'(k_2 a)\right]^2 \qquad (76)$$

VI. Conclusions

The results above indicate, first of all, that diffusion plays a very significant role in the saturation of molecular gases. Secondly, the saturation intensity in the case of inhomogeneous broadening is seen to be quite different from the saturation intensity in the case of homogeneous broadening. As a matter of fact, using equations (69) and (75), we can write

$$P_{out} - P_{in} = - \; [a^2 K'(k_2 a)] \quad A \qquad (77)$$

for the homogeneous case, and

$$\sqrt{P_{out}} - \sqrt{P_{in}} = - \; [aK'(k_2 a)] \quad B \qquad (78)$$

for the inhomogeneous case. A and B are constants.

The theoretical results derived herein were tested with seven molecular gases using various lines of the CO_2 laser around 10.6 microns. The gases were SF_6, BCl_3, C_2H_3Cl, NH_3, $C_2H_4F_2$, CH_2CF_2, and CCl_2F_2.

The most interesting observation during these experiments was that the output power did not vary much for constant input power when the radius of the beam was changed. Going back to equations (77) and (78), and taking this observation into consideration, we can say that if $a^2K'(k_2a)$ does not vary much when a is varied, the broadening should be homogeneous, and if $aK'(k_2a)$ does not vary much when a is varied, the broadening should be inhomogeneous. Table 2 is prepared for this purpose. It is seen that $aK'(k_2a)$ remains more or less constant in a large range of k_2a whereas $a^2K'(k_2a)$ increases quite fast with k_2a. Of course, we would expect $aK'(k_2a)$ to start increasing linearly with k_2a since $K'(k_2a)$ approaches 1 for very large values of k_2a.

The only way the function of $aK'(k_2a)$ can be constant with respect to a is that $K'(k_2a)$ varies inversely with a. From Table 3, we find

$$K'(k_2a) \doteq \frac{5.15}{a} \qquad (79)$$

in the range of k_2a indicated in Table 3. This result is most interesting. From a physical standpoint, the variation of the effect of diffusion inversely with the radius of the beam is intuitively sensible.

So if the inhomogeneous case is valid at low pressures, we can conclude that the variation of the output power with the radius of the beam at constant input power is negligible. We can write this conclusion in the form

TABLE 2

TEST FOR BROADENING

k_2a	$k_2a \cdot K'(k_2a)$	$(k_2a)^2 \cdot K'(k_2a)$
0.1	8.22	0.82
0.2	5.65	1.13
0.3	4.75	1.43
0.4	4.32	1.73
0.5	4.08	2.04
0.6	3.95	2.34
0.7	3.88	2.72
0.8	3.85	3.08
0.9	3.85	3.47
1.0	3.87	3.87
1.1	3.89	4.23
1.2	3.93	4.72
1.3	3.98	5.07
1.4	4.04	5.66
1.5	4.16	6.24
1.6	4.23	6.77
1.7	4.31	7.37
1.8	4.38	7.88
1.9	4.46	8.47
2.0	4.54	9.08
2.1	4.63	10.72
2.2	4.70	10.34
2.3	4.77	11.45
2.4	4.87	12.18
2.5	4.96	12.90
2.6	5.04	13.64
2.7	5.12	14.34
2.8	5.21	15.11
2.9	5.32	15.96
3.0	5.40	16.74
3.1	5.49	17.57
3.2	5.59	18.45
3.3	5.69	19.27
3.4	5.76	20.16
3.5	5.86	21.10
3.6	5.96	22.05
3.7	6.04	22.95
3.8	6.14	23.95
3.9	6.23	24.92

$$\frac{I_{out_1}}{I_{out_1}} \doteq \frac{a_2^2}{a_1^2} \qquad (80)$$

where the subscripts 1 and 2 refer to beam of radius a_1, and the beam of radius a_2. Equation (80) is expected to be valid at constant input power and low pressures. Using equations (70) and (76), we can also conclude that the saturation intensity varies inversely as the radius of the beam for the homogeneous broadening and varies inversely as the square of the radius of the beam in the case of inhomogeneous broadening, at low pressures and small radii.

VII. Appendix

Derivation of Effective Relaxation Rates and Diffusion Constants

Neglecting Diffusion, the rate equations can be written as[6]:

$$\dot{M}_2 = - w_2{}^u M_2 - \sum_i w_{2_i}^{u} M_2 + \sum_i w_{i_2}^{u} m_i^u - W(M_2 - M_1') + P_2{}^u \quad (A.1)$$

$$\dot{m}_k{}^u = - w_k{}^u m_k{}^u - \sum_i w_{k_i}^{u} m_k{}^u + \sum_i w_{ik}{}^u m_i{}^u + P_k{}^u \quad (A.2)$$

$$\dot{M}_1 = - w_1{}^1 M_1 - \sum_i w_{i1}{}^1 M_1 + \sum_i w_{i1}^{1} m_i + W(M_2 - M_i') + P_1{}^1 \quad (A.3)$$

$$\dot{m}_k{}^1 = - w_k{}^1 m_k{}^1 - \sum_i w_{k_i} m_k{}^1 + \sum_i w_{ik} m_i{}^1 + P_k{}^1 \quad (A.4)$$

In these equations various terms are defined as follows:

W = stimulated transition rate between upper and lower absorption levels.

$w_k{}^u$ = relaxation rate of the upper band kth level to all lower band levels

$P_k{}^u$ = pumping rate of the kth upper level

$P_k{}^1$ = pumping rate of the kth lower level

$w_k{}^1$ = relaxation rate of the lower band kth level to all other lower band levels

$w_{ki}^{u,1}$ = relaxation rate of the kth upper (lower) band level to the ith upper (lower) band level

$M_1' = \frac{g_2}{g_1} M_1$, where g_2 and g_1 are the degeneracies of levels 2 and 1

$m_k^{u,l}$ = population density of the upper(lower) band kth level

The number of equations and terms are actually infinite since k can assume any value. At steady state all the derivatives are zero. Adding equations (A.1) and (A.2), we get

$$0 = -\left[\frac{\sum_i w_i^u m_i^u}{M_2}\right] M_2 - \sum_i \sum_k w_{ki}^u m_k^u + \sum_i \sum_k w_{ik}^u m_k^u - W(M_2 - M_1') + \sum_k P_k^u \qquad (A.5)$$

The second and third terms cancel out. Assuming all the pumping rates are zero except for the upper absorption level, we obtain

$$\left[\frac{-\sum_i w_i^u m_i^u}{M_2}\right] M_2 - W(M_2 - M_1') + P_2^u = 0 \qquad (A.6)$$

Similarly, considering equations (A.3) and A.4), we get

$$\left[\frac{-\sum_i w_i^l m_i^l}{M_1}\right] M_1 + W(M_2 - M_1') + P_1^l = 0 \qquad (A.7)$$

We can assume all the rotational relaxation rates in each band are the same, equal to w^u and w^l. Then equations (A.6) and (A.7) are the same as the two-level model equations with the relaxation rate of the upper level replaced by $w^u \sum_i m_i^u / M_2$ and the relaxation rate of the lower level replaced by $w^l \sum_i m_i^l / M_1$.

In order to include the effect of diffusion in the rate equations, we have to add $D\nabla^2 M(\bar{r})$ term into equations (A.1) through (A.4). For example, equation (A.1) becomes

$$\dot{M}_2 = - w_2^u M_2 + D\nabla^2 M_2 - \sum_i w_{2i}^u M_2 + \sum_i w_{i2}^u m_i^u$$

$$- W(M_2 - M_1') + P_2^u \qquad (A.8)$$

Again assuming stead-state, if we add all the upper level equations, we obtain

$$- w_2 M_2 + D\nabla^2 \left(\frac{\sum_i m_i^u}{M_2}\right) M_2 - W(M_2 - M_1') + P_2^u = 0 \qquad (A.9)$$

So we can claim that the effective diffusion constant for the upper level D_2 is $D \sum_i m_i^u / M_2$. We obtain a similar result for the lower level by adding the lower level equations; namely, the effective diffusion constant for the lower level D_1 becomes $D \sum_i m_i^l / M_1$. Assuming that the rotational constant for the upper and lower bands are the same, D_1 and D_2 will be nearly equal.

REFERENCES

1. O.M. Stafsudd, O. Ersoy, S. Pizzica, Applied Optics, 10. 141. 1971.

2. O.R. Wood, S.E. Schwarz, Applied Physics Letters, 11, 88, 1967.

3. V. Ohtsuka, H. Yoshinaga, Japanese J. Applied Physics 8, 8, 1319, 1969.

4. T.Y. Chang, T.J. Bridges, Optical Communications, 1, 423, 1970.

5. J.P. Gordon, Bell Telephone Labs, Tech. Memo MM-63-1345-15, 1963.

6. O. Ersoy, UCLA Doctoral Dissertation, December 1972.

7. C.P. Christenson, C. Freed, H. A. Havs, IEEE J. Quantum Electronics, 5, 276, 1969.

8. O.R. Wood, P.L. Gordon, S.E. Schwarz, IEEE J. Quantum Electronics, 5, 502, 1969.

9. A. Yariv, Quantum Electronics, John Wiley and Sons, N.Y., 1967.