X. ELECTRODYNAMICS OF MEDIA[*]

Academic and Research Staff

Prof. L. J. Chu
Prof. H. A. Haus

Prof. P. Penfield, Jr.

Prof. T. J. Bridges
J-P. E. Taran

Graduate Students

C. P. Christensen, Jr.
H. Granek

K. T. Gustafson
P. W. Hoff
E. R. Kellett, Jr.

J. T. Patterson
C. M. Watson

A. SATURATION POWER IN CO_2 LASERS

If we apply the formula for the saturation power of a two-level system[1] to the case of CO_2, using for the relaxation time of the upper level the value 10^{-3} sec, for the lower level 10^{-4}, and $t_{sp} = 5$ sec, we obtain a value that is too small compared with the experimentally observed value by a factor of 500 or 100, the value depending on the experiment that is used for comparison. Bridges and Kogelnik[2] quote 100 w/cm^2, Miles[3] quotes 60 w/cm^2, and Hotz[4] quotes 22 w/cm^2.

Since the CO_2 laser is not a two-level system, in that the different J levels relax into the lasing vibrational-rotational transition, it may be argued that many levels participate in the lasing action. Hotz and Austin,[4] have tried to explain the discrepancy by assuming that 50 levels participate. This is an unreasonably large number of levels.

In this report, we determine the cross relaxation of the rotational levels, by solving in closed form the rate equations in the limit when the vibrational relaxation times are long compared with the rotational relaxational times, a situation that holds in practice. It may be shown that the number of levels participating in CO_2 laser action is of the order of 15. Hence the discrepancy between the theoretically predicted value for the saturation power and the one observed experimentally cannot be explained solely by assuming cross relaxation of the rotational levels. We propose that the discrepancy can be explained by further assuming spatial diffusion of the unused populations into the laser beam, and we estimate the magnitude of this effect.

We denote the number density of particles in the kth rotational level of the upper $00°1$ vibrational level by N_k, and the population density of the jth rotational level in the lower $10°0$ vibrational level by n_k. The rate equations are then

$$\frac{dN_1}{dt} = R_1^u - N_1\gamma_1^u - \sum_j \Gamma_{j1}^u N_1 + \sum_j \Gamma_{1j}^u N_j - W(N_1 - gn_1)$$

$$\frac{dN_2}{dt} = R_2^u - N_2\gamma_2^u - \sum_j \Gamma_{j2}^u N_2 + \sum_j \Gamma_{2j}^u N_j \tag{1}$$

. . .

$$\frac{dn_1}{dt} = r_1^\ell - n_1\gamma_1^\ell - \sum_j \Gamma_{j1}^\ell n_1 + \sum_j \Gamma_{1j}^\ell n_j + W(N_1 - gn_1)$$

$$\frac{dn_2}{dt} = r_2^\ell - n_2\gamma_2^\ell - \sum_j \Gamma_{j2}^\ell n_2 + \sum_j \Gamma_{2j}^\ell n_j \tag{2}$$

. . . .

Here the quantities R_k^u and r_j^ℓ give the rate of pumping into the kth upper rotational level and j^{th} lower rotational level, respectively. The vibrational relaxation constants γ_k^u may be assumed to be different in all levels, but in general are much smaller than the constants of cross relaxation Γ_{jk}. The superscripts u and ℓ denote the upper and lower vibrational levels. The rate of induced transitions is given by

$$W = \frac{\lambda^2 I}{8\pi^2 t_{sp} h\nu\Delta\nu} \tag{3}$$

where I is the intensity of the lasing radiation, and t_{sp} is the spontaneous transition time. In order to find a simple expression for the gain, we define the determinants

$$\Delta^u = \begin{vmatrix} -\left(\gamma_1^u + \sum_j \Gamma_{j1}^u\right) & \Gamma_{12}^u & \Gamma_{13}^u & \cdots & \Gamma_{1M}^u \\ \Gamma_{21}^u & -\left(\gamma_2^u + \sum_j \Gamma_{j2}^u\right) & \Gamma_{23}^u & \cdots & \Gamma_{2M}^u \\ \Gamma_{31}^u & \Gamma_{32}^u & -\left(\gamma_3^u + \sum_j \Gamma_{j3}^u\right) & \cdots & \Gamma_{3M}^u \\ \vdots & \vdots & \vdots & & \\ \Gamma_{M1}^u & \Gamma_{M2}^u & \Gamma_{M3}^u & \cdots & -\left(\gamma_M^u + \sum_j \Gamma_{jM}^u\right) \end{vmatrix} \tag{4}$$

and

$$
A^u = \begin{vmatrix}
-\left(\gamma_2^u + \sum_j \Gamma_{j2}^u\right) & \Gamma_{23}^u & \Gamma_{24}^u & \cdots & \Gamma_{2M}^u \\
\Gamma_{32}^u & -\left(\gamma_3^u + \sum_j \Gamma_{j3}^u\right) & \Gamma_{34}^u & \cdots & \Gamma_{3M}^u \\
\Gamma_{42}^u & \Gamma_{43}^u & -\left(\gamma_4^u + \sum_j \Gamma_{j4}^u\right) & \cdots & \Gamma_{4M}^u \\
\vdots & \vdots & \vdots & & \\
\Gamma_{M2}^u & \Gamma_{M3}^u & \Gamma_{M4}^u & \cdots & -\left(\gamma_M^u + \sum_j \Gamma_{jM}^u\right)
\end{vmatrix}
$$

(5)

Δ^u is the determinant describing the relaxation among the upper rotational levels in the absence of a lasing field. A^u is the subdeterminant obtained by removing the first row and column from Δ^u. Corresponding quantities can be defined for the lower level. If we define the quantity K_u by the determinant obtained by replacing the first column of Δ^u by the pumping rates and introduce a corresponding quantity K_ℓ for the lower set of levels, we finally have the weighted difference between the populations, which is proportional to the gain:

$$
N_1 - gn_1 = \frac{-K_u \Delta^\ell + gK_e \Delta^u}{\Delta^u \Delta^\ell - W(A^u \Delta^\ell + gA^\ell \Delta^u)}
$$

(6)

The lasing power appears only in the denominator. The saturation power is defined by that value of intensity which reduces the right-hand side of (6) to one-half its value for zero lasing power.

$$
P_s = \frac{8\pi^2 t_{sp} h\nu\Delta\nu}{\lambda^2} \left[\frac{1}{-\dfrac{A^u}{\Delta^u} - \dfrac{A^\ell}{\Delta^\ell} g} \right].
$$

(7)

We shall now evaluate the saturation power in the limit when the vibrational relaxation rates are slow compared with the rotational cross-relaxation rates. Expanding the determinant Δ^u to first order in the vibrational relaxation rate, we have

$$
\Delta^u = -\sum_k \gamma_k^u \beta_k,
$$

(8)

where β_k^u is the cofactor of the kth diagonal term, with γ_j^u set equal to 0 for

all j. From the principle of detailed balance, we have

$$N_j^e \Gamma_{ij}^u = N_i^e \Gamma_{ji}^u. \tag{9}$$

Using this fact and dividing all columns by their corresponding equilibrium number density, we can show that the cofactors are all proportional to a common number β_o^u.

$$\beta_j^u = \frac{N_j^e}{\prod\limits_k N_k^e} \beta_o^u. \tag{10}$$

Now note that since the determinants Δ^u and Δ^ℓ are of first order in the vibrational relaxational times, we need not go farther than zero order to evaluate A^u and A^ℓ. These to zero order are

$$A^u = \frac{N_1^e}{\prod\limits_k N_k^e} \beta_o^u. \tag{11}$$

When these expressions are introduced into the saturation power density, we obtain

$$P_s = \frac{8\pi^2 t_{sp} h\nu\Delta\nu}{\lambda^2} \left[\frac{\dfrac{\sum\limits_i \gamma_i^u N_i^e}{N_1^e} \quad \dfrac{\sum\limits_i \gamma_j^\ell n_j^e}{n_1^e}}{g\dfrac{\sum\limits_i \gamma_i^u N_i^e}{N_1^e} + \dfrac{\sum\limits_j \gamma_j^\ell n_j^e}{n_1^e}} \right]. \tag{12}$$

Assuming $\gamma_i^u = \gamma^u$ and $\gamma_j^e = \gamma^e$, namely that all relaxation rates within one level are the same, we find that the saturation power density contains the vibrational relaxation times multiplied by an effective number of upper and lower levels.

$$n_1' = \frac{\sum\limits_{k \text{ even}}^{\infty} n_k^e}{n_1^e} \tag{13}$$

$$N_1' = \frac{\sum\limits_{k \text{ odd}}^{\infty} N_k^e}{N_1^e}. \tag{14}$$

How large are these? Considering the lower level as an example, we have

$$n'_1 = \frac{\sum\limits_{J\ \text{even}}^{\infty} (2J+1) \exp\left(-BJ(J+1)\frac{hc}{KT}\right)}{(2J_1+1) \exp\left(-BJ_1(J_1+1)\frac{hc}{KT}\right)}.$$ (15)

We may approximate the summations by integrations, noting, however, that the lower level has only even terms. In this way one obtains

$$n'_1 = \frac{KT}{2Bhc(2J_1+1) \exp\left(-BJ_1(J_1+1)\frac{hc}{KT}\right)}.$$ (16)

Assume next that the level of interest is the one with the largest population density. The J number of this level is given by

$$\frac{Bhc}{KT} = \frac{2}{(2J_1+1)^2}.$$

When we introduce into these expressions the corresponding numbers for CO_2, we find

$$n'_1 = 14.3$$ (17)

$$N'_1 = 14.7.$$ (18)

Hence, only 15 or so levels are participating in the lasing action. Introducing the previously mentioned values for γ^u and γ^ℓ into Eq. 12, we obtain for the saturation power

$$P_s = 5.8 \text{ w/cm}^2.$$ (19)

This value is still considerably smaller than the experimentally observed values. We have to look, therefore, for other mechanisms that could account for the observations.

The problem of solving for spatial diffusion of the population densities in the various rotational levels under the influence of a lasing field is prohibitively difficult. One may make some reasonable approximations and assumptions, however, which lead to an estimate of the magnitude of this effect, if the rotational levels are coupled very tightly to the lasing level. Their population densities are reduced in the upper level and increased in the lower level, in the ratio of their equilibrium densities. We shall also assume, for simplification, that only 15 levels participate, all of which have the same population density. Furthermore, we shall consider a case of a very thin laser beam of uniform intensity, so that we may assume that the population densities within the laser beam are spatially uniform. Under such an assumption, we need to solve the diffusion problem only in the region surrounding

the laser beam. As the population of the upper vibrational level decreases within the laser beam, the particles situated outside the beam diffuse into the beam and partially compensate for the decrease. The diffusion of a single species is governed by the equation

$$D \nabla^2 N - \gamma^u N + R^u = 0,$$ (20)

where D is the diffusion constant (cm^2/sec). The solution of this equation is a modified Bessel function having the asymptotic behavior

$$K_0(x) \approx \frac{2}{\pi} \ln \frac{2}{\gamma x}.$$ (21)

The rate at which particles are supplied to the core is given by

$$-D \frac{\partial N}{\partial r} = -D \left[\frac{N - \frac{R^u}{\gamma^u}}{\frac{2}{\pi} \ln \frac{2}{\gamma \rho \sqrt{\gamma^u/D}}} \right] \frac{2}{\pi r},$$ (22)

which leads to an equivalent source S per unit volume that should be added to the rate equation of the particles inside the laser beam:

$$S = -D \left[\frac{N - \frac{R^u}{\gamma^u}}{\ln \frac{2}{\gamma \rho \sqrt{\gamma^u/D}}} \right] \frac{2}{\rho^2}.$$ (23)

We see that the effect of diffusion in this simple case is to supplement the rate constant by vibrational relaxations, thereby replacing it by

$$\gamma^{u'} = \gamma^u + \frac{2D}{\rho^2} \frac{1}{\ln \frac{2}{\gamma \rho \sqrt{\gamma^u/D}}}.$$ (24)

Furthermore, the source too is modified:

$$R^{u'} = R^u \left[1 + \frac{2D}{\rho^2} \frac{1}{\gamma^u \ln \frac{2}{\gamma \rho \sqrt{\gamma^u/D}}} \right].$$ (25)

The main interest is in the relaxation constant, however, because the saturation parameter depends upon the relaxation constant and not on the pumping rate. When reasonable numbers and beam diameters are introduced into these expressions, we

find for the net effective vibrational relaxation rate of the upper level

$$\gamma^{u'} = 3.5 \times 10^3 \text{ sec}^{-1}, \tag{26}$$

and of the lower level

$$\gamma^{\ell'} = 1.25 \times 10^4 \text{ sec}^{-1}. \tag{27}$$

When the geometry of the Hotz experiment is taken into account, we find

$$P_s = 14 \text{ w/cm}^2. \tag{28}$$

For Bridges' experiment, we find

$$P_s = 21 \text{ w/cm}^2. \tag{29}$$

We note that the spatial diffusion changes appreciably the saturation parameter from the one that was obtained when diffusion is disregarded. In the past, diffusion was not considered an important contributor to the saturation parameter. This was mainly due to the fact that atomic and ionic lasers have relaxation rates of the lasing levels that are considerably larger than those of molecular lasers. In those cases, diffusion rates are certainly negligible. On the other hand, the relaxation rates of the vibrational levels for CO_2 are relatively low, and hence diffusion can play an important role. This will be true presumably for other molecular lasers as well.

C. P. Christensen, Jr., H. A. Haus

References

1. J. P. Gordon, Bell Telephone Laboratories Memorandum MM-63-1353-15, 1963 (unpublished).
2. H. Kogelnik and T. J. Bridges, IEEE J. Quantum Electronics, Vol. QE-3, p. 95, 1967.
3. P. A. Miles and J. W. Lotus, a paper presented at the 1968 International Quantum Electronics Conference.
4. D. F. Hotz and J. W. Austin, Appl. Phys. Letters 11, 60 (1967).
5. W. W. Rigrod, J. Appl. Phys. 34, 2602 (1963).

B. THEORY AND EXPERIMENT OF ROTATIONAL CROSS RELAXATION IN CO_2 LASER

1. Introduction

A study of the coupling between the rotational lines of two lasing vibrational levels in the CO_2 laser has been undertaken in an effort to determine the rotational relaxation

times. This project involves passing a low-power laser that is tunable to several J transitions through a five-pass laser amplifier simultaneously with a high-power laser that is tunable to a single (different) line, and observing the gain profile as a function of the power of the single line.

2. Theory

The time variation of the population densities of the set of quantum levels are described[1] as

$$\dot{\rho}_{\lambda\lambda}(t) = \sum_{\nu} \rho_{\nu\lambda}(t)\, \Gamma_{\nu\lambda} - \sum_{\nu} \rho_{\lambda\lambda}(t)\, \Gamma_{\lambda\nu},$$ (1)

where $\rho_{\nu\lambda}(t)$ is the respective density matrix element. Conservation of energy requires

$$\rho^e_{\nu\nu}\Gamma_{\nu\lambda} = \rho^e_{\lambda\lambda}\Gamma_{\lambda\nu}.$$ (2)

Multiplying (1) and (2) by the total number of molecules present yields in terms of the total number density

$$\dot{N}_k(t) = \sum_{i} N_i(t)\, \Gamma_{ik} - \sum_{i} N_k(t)\, \Gamma_{ki} + R_k$$ (3)

$$N^e_\lambda \Gamma_{\lambda\nu} = N^e_\nu \Gamma_{\nu\lambda},$$ (4)

where a source term R_k external to the subsystem that is being considered has been included. Furthermore, in this treatment we shall incorporate a saturating term on the saturating line that will have a dependence on the nondegenerate population inversion of that level, while all other terms will be determined solely by the relaxation among themselves. In equilibrium the left-hand side of (3) is zero, and by considering K levels in addition to the saturating level in each of the upper and lower vibrational levels, this analysis yields a set of equations:

$$-N^u_s\left(\Gamma^u_s + \sum_{k} \Gamma^u_{sk}\right) + \sum_{k} \Gamma^u_{ks}N^u_k - P\left(\frac{N^u_s}{g^u_s} - \frac{N^\ell_s}{g^\ell_s}\right) = -R^u_s$$ (5)

$$N^u_s\Gamma^u_{sk'} - N^u_{k'}\left[\Gamma^u_{k'} + \Gamma^u_{ks} + \sum_{k\neq k'} \Gamma^u_{k'k}\right] + \sum_{k\neq k'}\left(N^u_{k'}\Gamma^u_{k'k}\right) = -R^u_{k'}$$ (6)

$$-N_s^\ell \left(\Gamma_s^\ell + \sum_k \Gamma_{sk}^\ell \right) + \sum_k \Gamma_{ks}^\ell N_k^\ell + P \left(\frac{N_s^u}{g_s^u} - \frac{N_s^\ell}{g_s^\ell} \right) = -R_s^\ell \tag{7}$$

$$N_s^\ell \Gamma_{sk'}^\ell - N_{k'}^\ell \left[\Gamma_{k'}^\ell + \Gamma_{ks}^\ell + \sum_{k \neq k'} \Gamma_{k'k}^\ell \right] + \sum_{k \neq k'} \left(N_{k'}^\ell \Gamma_{k'k}^\ell \right) = -R_{k'}^\ell \tag{8}$$

where the g are the appropriate degeneracy factors. Also, the equilibrium densities and the relaxation rates obey the relations

$$N_{\lambda_e}^u \Gamma_{\lambda\nu}^u = N_{\nu_e}^u \Gamma_{\nu\lambda}^u = \gamma^u$$

$$N_{\lambda_e}^\ell \Gamma_{\lambda\nu}^\ell = N_{\nu_e}^\ell \Gamma_{\nu\lambda}^\ell = \gamma^\ell \tag{9}$$

$$N_{\lambda_e}^u \Gamma_\lambda^u = a^u$$

$$N_{\lambda_e}^\ell \Gamma_\lambda^\ell = a^\ell, \tag{10}$$

where the first equality of (9) follows from (4) and (10), and the last equality of (9) follows from the assumption that all rotational levels in a vibrational state relax to equilibrium under identical collision conditions.

Solving the K equations represented by (6) in terms of N_s^u yields a matrix for the upper state of the form

$$\begin{bmatrix} -\left[\Gamma_1^u + \sum\limits_{\substack{k \neq 1 \\ s}} \Gamma_{1k}^u\right] & \Gamma_{21}^u & \Gamma_{31}^u \cdots & \Gamma_{K1}^u \\ \Gamma_{12}^u & -\left[\Gamma_2^u + \sum\limits_{\substack{k \neq 2 \\ s}} \Gamma_{2k}^u\right] & & \vdots \\ \Gamma_{1K}^u & \Gamma_{2K}^u & \cdots & -\left[\Gamma_K^u + \sum\limits_{\substack{k \neq K \\ s}} \Gamma_{KK}^u\right] \end{bmatrix} \begin{bmatrix} N_1^u \\ N_2^u \\ N_K^u \end{bmatrix} = \begin{bmatrix} -N_s^u \Gamma_{s1}^u - R_1 \\ -N_s^u \Gamma_{s2}^u - R_2 \\ -N_s^u \Gamma_{sK}^u - R_K \end{bmatrix} \tag{11}$$

Multiplying all of the terms in the first column by N_1^e/N_1^e, in the second column by N_2^e/N_2^e, and so forth reduces (11) to

$$
\begin{bmatrix}
-\left(\dfrac{a^u + K\gamma^u}{N_1^e}\right) & \dfrac{\gamma^u}{N_2^e} & \dfrac{\gamma^u}{N_3^e} & \cdots & \dfrac{\gamma^u}{N_K^e} \\[2ex]
\dfrac{\gamma^u}{N_1^e} & -\left(\dfrac{a^u + K\gamma^u}{N_2^e}\right) & \cdots & & \\[2ex]
\dfrac{\gamma^u}{N_1^e} & & \cdots & & \\[2ex]
\cdots & & & & \\[2ex]
\dfrac{\gamma^u}{N_1^e} & & & & -\left(\dfrac{a^u + K\gamma^u}{N_k^e}\right)
\end{bmatrix}
\begin{bmatrix}
N_1 \\[2ex] N_2 \\[2ex] \\[2ex] \\[2ex] N_K
\end{bmatrix}
=
\begin{bmatrix}
-\dfrac{N_s}{N_s^e}\gamma^u - R_1 \\[2ex]
-\dfrac{N_s}{N_s^e}\gamma^u - R_2 \\[2ex] \\[2ex] \\[2ex]
-\dfrac{N_s}{N_s^e}\gamma^u - R_k
\end{bmatrix}
$$

$$(12)$$

By Cramér's rule, (12) can be solved for each of the N_k.

$$
N_k = N_k^e \left[\frac{N_s}{N_s^e} \, \frac{1}{\left(\frac{a}{\gamma}+1\right)} + \frac{R_k\left(\frac{a}{\gamma}+1\right) + \sum\limits_{i=1}^{K} R_i}{\gamma\left(\frac{a}{\gamma}+1\right)\left(\frac{a}{\gamma}+K+1\right)} \right].
\tag{13}
$$

By a study of the equilibrium equations, it can be seen that it is necessary to set the pumping rates, R_k^u, equal to each other, $R_k^u = R_o^u$, and likewise for the lower vibrational level. Under this assumption, we have

$$
N_k^u = N_{k_e}^u \left[\frac{N_s^u}{N_{s_o}^u} \, \frac{1}{\beta^u + 1} + \frac{R_o^u}{\gamma^u(\beta^u+1)} \right].
\tag{14}
$$

Here $N_{s_o}^u$ is the equilibrium density of the upper saturating level, and

$$
\beta^u = \frac{a^u}{\gamma^u}.
\tag{15}
$$

After modifying (5),

$$
-\frac{N_s^u}{N_{s_o}^u}(a^u + K\gamma^u) + \gamma^u \sum_k \frac{N_k^u}{N_{k_e}^u} - P\left(\frac{N_s^u}{g_s^u} - \frac{N_s^\ell}{g_s^\ell}\right) = -R_s^u,
\tag{16}
$$

and inserting (14) into (16), we attain for the upper state

$$-\left[\frac{P}{g_s^u} + \frac{\gamma^u}{N_{s_o}^u}\left(\beta^u + K - \frac{K}{\beta^u + 1}\right)\right] N_s^u + \frac{P}{g_s^\ell} N_s^\ell = -R_o^u\left(\frac{K}{\beta^u + 1} + 1\right), \tag{17}$$

and similarly for the lower state

$$\frac{P}{g_s^u} N_s^u - \left[\frac{\lambda^u}{N_{s_o}^u}\left(\beta^\ell + K - \frac{K}{\beta^\ell + 1}\right) + \frac{P}{g_s^\ell}\right] N_s^\ell = -R_o^\ell\left(\frac{K}{\beta^\ell + 1}\right). \tag{18}$$

Defining

$$T^{u,\ell} = \beta^{u,\ell} + K - \frac{K}{\beta^{u,\ell} + 1} \tag{19}$$

and simultaneously solving (17) and (18), yields

$$N_s^u = \frac{R_o^u\left[1 + \frac{K}{\beta^u + 1}\right]\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell} + \frac{P}{g_s^\ell}\left[R_o^u\left(1 + \frac{K}{\beta^u + 1}\right) + R_o^\ell\left(1 + \frac{K}{\beta^\ell + 1}\right)\right]}{\frac{\gamma^u T^u}{N_{s_o}^u}\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell} + \left(\frac{\gamma^u T^u}{N_{s_o}^u g_s^\ell} + \frac{\gamma^\ell T^\ell}{N_{s_o}^\ell g_s^u}\right)P} \tag{20}$$

$$N_s^\ell = \frac{R_o^\ell\left[1 + \frac{K}{\beta^\ell + 1}\right]\frac{\gamma^u T_k^u}{N_{s_o}^u} + \frac{P}{g_s^u}\left[R_o^\ell\left(1 + \frac{K}{\beta^\ell + 1}\right) + R_o^u\left(1 + \frac{K}{\beta^u + 1}\right)\right]}{\frac{\gamma^u T^u}{N_{s_o}^u}\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell} + \left(\frac{\gamma^u T^u}{N_{s_o}^u g_s^\ell}\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell g_s^u}\right)P}. \tag{21}$$

The net gain coefficient for a rotational transition between the upper vibrational level, 1, and the lower vibrational level, 2, is given[2] by

$$a_{1_{J-1}2_J} = \left(\frac{\ln 2}{\pi}\right)^{1/2}\frac{16\pi^3 c^3}{3h\Delta\nu_D \lambda_{1_{J-1}2_J}}[JK_{12}]\left[\frac{N_{1_{J-1}}}{g_{J-1}} - \frac{N_{2_J}}{g_J}\right] \tag{22}$$

for a Doppler-broadened laser, where JK_{12} is the matrix element of the transition, J being the lower state rotational quantum number,

$$\Delta\nu_D = \frac{2}{\lambda_{1_{J-1}2_J}}\left[\left(\frac{2kT}{M}\right)\ln 2\right]^{1/2},\tag{23}$$

the degeneracy factor $g_J = (2J+1)$, and M is the molecular mass. Letting $N_s^\ell = N_{2_{J_o}}$

and $N_s^u = N_{1_{J_o-1}}$ and using (22) and (23), we have an expression for the P-transition gain

on the saturating line, J_o,

$$a_{1_{J_o-1}2_{J_o}} = (Z)\frac{\frac{J_o}{(2J_o-1)}R_o^u\left(1+\frac{K}{\beta^u+1}\right)\frac{\gamma^\ell T_k^\ell}{N_{s_o}^\ell}-R_o^\ell\left(1+\frac{K}{\beta^u+1}\right)\frac{\gamma^u T_k^u}{N_{s_o}^u}\frac{J_o}{(2J_o+1)}}{\frac{\gamma^u T_k^u}{N_{s_o}^u}\frac{\gamma^\ell T_k^\ell}{N_{s_o}^\ell}+\left(\frac{\gamma^u T_k^u}{N_{s_o}^u g_s^\ell}+\frac{\gamma^\ell T_k^\ell}{N_{s_o}^\ell g_s^u}\right)P}.\tag{24}$$

By the same notation, if $N_k^\ell = N_{2_J}$ and $N_k^u = N_{1_{J-1}}$, the gain of the P(J) transition is

$$a_{1_{J-1}2_J} = JZ\left(\frac{N_k^u}{g_{J-1}}-\frac{N_k^\ell}{g_J}\right) =$$

$$(Z)\left[\frac{\left[R_o^u\left(1+\frac{K}{\beta^u+1}\right)+R_o^\ell\left(1+\frac{K}{\beta^\ell+1}\right)\right]\left[\frac{2J_o-1}{(1+\beta^u)(2J-1)}\frac{N_{e_{J-1}}^u}{N_{e_{J_o-1}}^u}-\frac{2J_o+1}{(1+\beta^\ell)(2J+1)}\frac{N_{e_J}^\ell}{N_{e_{J_o}}^\ell}\right]\frac{JP}{4J_o^2-1}}{\frac{\gamma^u T^u}{N_{s_o}^u}\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell}+\left(\frac{\gamma^u T^u}{N_{s_o}^u g_s^\ell}+\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell g_s^u}\right)P}\right.$$

$$+\frac{\left[\frac{N_{e_{J-1}}^u}{N_{e_{J_o-1}}^u}\frac{1}{2J-1}R_o^u\left(\frac{K}{\beta^u+1}\right)\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell}\frac{1}{(\beta^u+1)}-\frac{N_{e_J}^\ell}{N_{e_{J_o}}^\ell}\frac{1}{2J+1}R_o^\ell\left(\frac{K}{\beta^\ell+1}\right)\frac{\gamma^u T^u}{N_{s_o}^u}\left(\frac{1}{\beta^\ell+1}\right)\right]J}{\frac{\gamma^u T^u}{N_{s_o}^u}\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell}+\left(\frac{\gamma^u T^u}{N_{s_o}^u g_s^\ell}+\frac{\gamma^\ell T^\ell}{N_{s_o}^\ell g_s^u}\right)P}$$

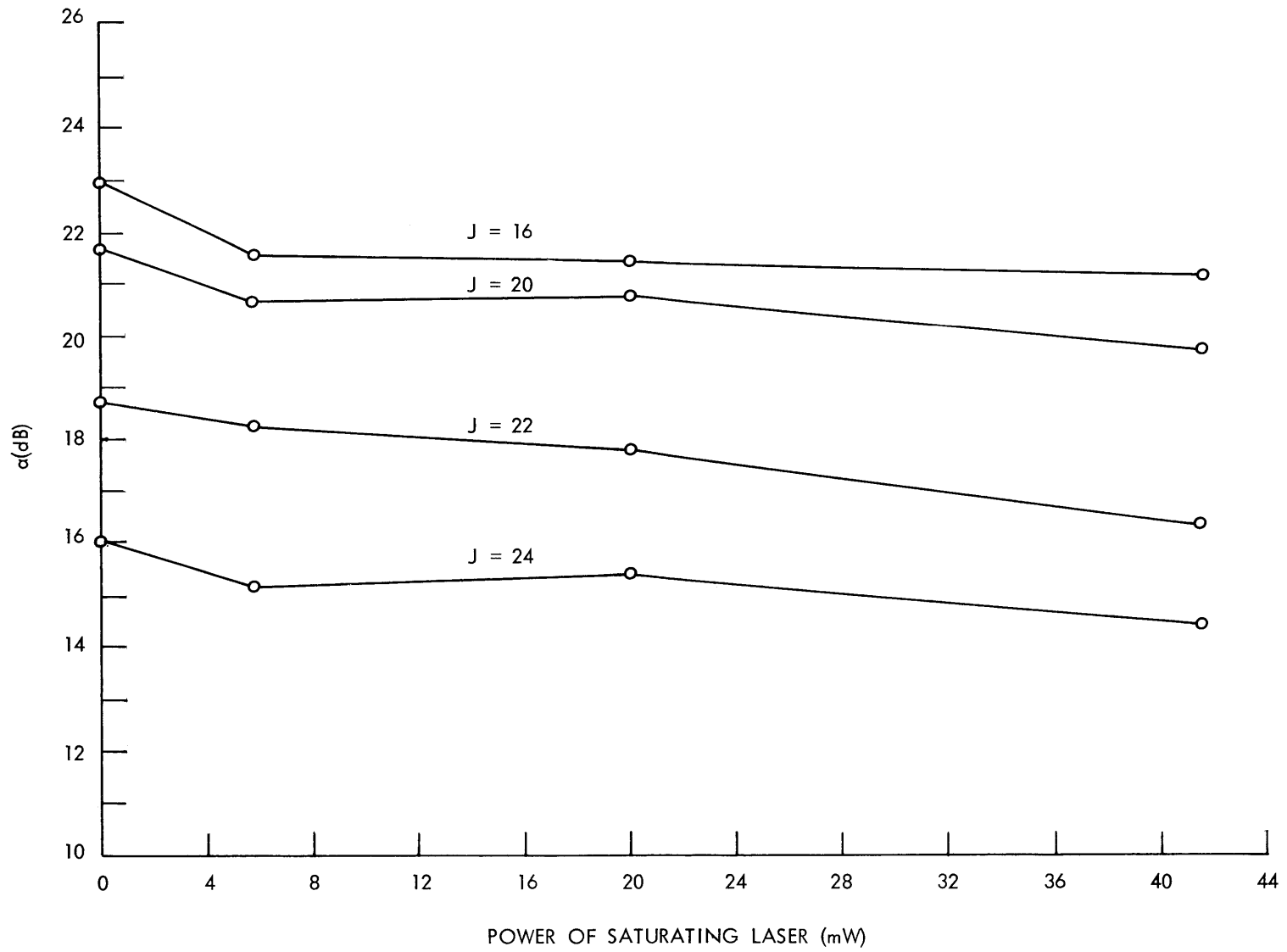$$\left.+\left[\frac{R_o^u}{\gamma^u(\beta^u+1)}\frac{J}{2J-1}-\frac{R_o^\ell}{\gamma^\ell(\beta^\ell+1)}\frac{J}{2J+1}\right]\right],\tag{25}$$

Fig. X-1.   Gain vs saturating power of P(18).

where

$$Z = \frac{8\pi^3 c^3}{3h} \left[\frac{M}{2kT}\right]^{1/2} K_{12},$$

and P is related to the incident flux density, I, by

$$P = \frac{\lambda^3}{8\pi hc} \frac{1}{t_{spont}} \frac{I}{2\pi\Delta\omega}. \tag{26}$$

Taking the limit as P goes to infinity and looking at the ratio of gains on the Jth line to that on the Lth line results in

$$\frac{a_{12}(J)}{a_{12}(L)} = \frac{\frac{1}{1+\beta^u}\exp\left[B_u\frac{hc}{kT}\left(\left(J_o^2-J^2\right)-(J_o-J)\right)\right] - \frac{1}{1+\beta^u}\exp\left[B_\ell\frac{hc}{kT}\left(\left(J_o^2-J^2\right)+(J_o-J)\right)\right]}{\frac{1}{1+\beta^u}\exp\left[B_u\frac{hc}{kT}\left(\left(J_o^2-L^2\right)-(J_o-L)\right)\right] - \frac{1}{1+\beta^u}\exp\left[B_\ell\frac{hc}{kT}\left(\left(J_o^2-L^2\right)+(J_o-L)\right)\right]}\frac{J}{L}, \tag{27}$$

where all terms are determinable except $\gamma_{rot} = \frac{\beta^\ell}{a^\ell} = \frac{\beta^u}{a^u}$, under the assumption that the rotation relaxation rate is the same for the upper and lower levels. With increasing saturating power, the gain on each line decreases in approximately the same fashion, until it asymptotically reaches a constant as the power goes to infinity. From an experimental plot of gain against power for the different P transitions we can thereby determine the inverse rotational relaxation time, γ, since all other parameters in (27) are determinable from other experiments or are known physical constants.

3. Experimental Results

With the high-power laser on the P(18) transition and its power varied by means of a pair of polarizers, and the probe laser tunable to P(16), P(20), P(22), and P(24) the gain versus saturating power curve (Fig. X-1) was obtained. From a comparison of the leveling-off gains of the different P transitions we have determined[3] a τ_{rot} of between 60 nsec and 180 nsec, for a $\tau_{vib}^u = 1$ msec $\tau_{vib}^\ell = 0.1$ msec. The variation has probably been most severely affected by the failure of the assumption that we were sufficiently saturating P(18) to have reached gains on the other lines relatively independent of the saturating power.

H. Granek

References

1. A. A. Vuylsteke, Maser Theory (D. Van Nostrand Company, Inc., Princeton, N. J., 1960).
2. C. K. N. Patel, "Interpretation of CO_2 Optical Maser Experiments," Phys. Rev. Letters 12, 21 (May 25, 1964).
3. C. G. B. Garrett, Gas Lasers (McGraw-Hill Publishing Company, New York, 1967).