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TIME DEPENDENT PULSE AMPLIFICATION IN A THREE LEVEL GAS 1/1
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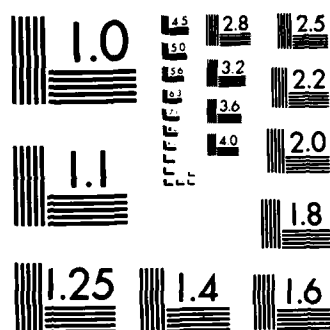
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Time Dependent Pulse Amplifications in a three Level Gas

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PROGRESS REPORT

to

Air Force Office of Scientific Research

from

Professor T. F. Morse
Division of Engineering
Brown University
Providence, RI 02912

January 1, 1980 - December 31, 1980

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SCIENTIFIC RESULTS AND PROGRESS DURING THE PAST 12 MONTHS

THEORETICAL

The main theoretical results of our past year's work is contained in the paper with T. Tsai, 'Time Dependent Pulse Amplification in a Three Level Gas'. The abstract is given below.

→ The time dependent density matrix equations have been solved in conjunction with the field amplitude equations for a three level optically pumped gas. The solutions are valid in the limit in which the pumping pulse is longer than the dephasing time of the system and true coherent effects may be neglected. It is assumed that the FIR field is smaller than the pump field, and saturation effects associated with this field are absent. In the limit in which the pump pulse is resonant, particularly simple results for the FIR field amplification may be obtained. The solutions exhibit the dependence of the FIR output pulse on the shape of the input pulse. Further, as a consequence of non-linear saturation effects on the pump pulse transition, the FIR pulse is initially created with two peaks, one associated with the leading edge and one with the trailing edge of the pump pulse. As amplification on the FIR transition and pump absorption increase, these two peaks merge. ←

Enclosed is a copy of the paper that has been forwarded to the IEEE Journal of Quantum Electronics.

EXPERIMENTAL

Experimental work over the past year has been concerned with the detailed measurement of breakdown characteristics of argon gas. It had been hoped that by measuring the time dependence of the breakdown in a gas, that certain 'finger-prints' would appear that independent of the statistical

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nature of laser induced gas breakdown. Several interesting aspects of laser induced gas breakdown were observed, some of which were not reported previously in the literature, and it was learned that the only characteristic we could isolate that did not vary statistically, was the slope of beam attenuating plasma as a function of pressure. It had been hoped that this pressure dependence would be sensitive enough for this technique to be employed as a diagnostic tool; however, this does not appear to be the case. In addition to obtaining the first laser induced gas breakdown in argon in which the temporal behavior of the breakdown was monitored, this was done with a single mode laser so that a more accurate value of threshold could be obtained. These results are being prepared for publication in the Journal of Applied Physics. In addition, we have discovered a relatively simple method to measure the divergence angle of a carbon dioxide laser beam using burn patterns on uncoated high speed polaroid film. This depends upon the existence of a gaussian beam, and upon the distinct two level sensitivity of the film to the burn pattern. We hope to prepare these useful results in a note.

PREVIOUS ACCOMPLISHMENTS

Previous accomplishments have been associated with the development of kinetic equations for polyatomic gases, and the solution of various boundary value problems of specific

relevance to the space program. More recent work has been concerned with the interaction of radiation and gases in a three level system, and equations have been derived from the density matrix that are rate equations retaining nonlinear power broadening and multiphoton effects. These equations have been solved in the case in which the pumping field is stronger than the FIR field, and such solutions should be of interest to experimentalists concerned with the optimization of of FIR optically pumped lasers.

PUBLICATIONS

1. Time Dependent Pulse Amplification in a Three Level Gas, with T.Tsai, submitted to IEEE J. Quant. Electron.

OTHER SOURCES OF SUPPORT

During the past year, support was solely through the Air Force Office of Scientific Research.

PERSONNEL

1. Professor T.F. Morse, Division of Engineering, Brown University, Principal Investigator.
2. Eugene Potkay, graduate student engaged in the experimental part of the project. He has completed his dissertation and is employed at Western Electric.
3. T. Tsai, graduate student, completed his dissertation and is presently employed in a consulting firm in the Washington area.

Time Dependent Pulse Amplification in a Three Level Gas

by

Tsung-Ming Tsai and T.F. Morse

Division of Engineering, Brown University,
Providence, R.I. 02912

ABSTRACT

The density matrix equations have been solved in conjunction with the time dependent field amplitude equations for a three level optically pumped gas. The solutions are valid in the limit in which the pumping pulse is longer than the dephasing time of the system and true coherence effects may be neglected. In order to obtain closed form solutions, it is assumed that the FIR field is smaller than the pump field and saturation effects associated with this field are absent. When the pump pulse is resonant with the absorbing FIR transition, particularly simple solutions for the time dependent FIR field amplification are obtained. These solutions exhibit the specific dependence of the FIR output pulse on the shape of the input pulse and display certain pulse narrowing features.

1. INTRODUCTION

There has been much recent interest in both experimental as well as theoretical studies of optically pumped gas laser systems, with particular emphasis on FIR wavelengths.¹⁻¹³ An excellent review is presented in reference 14. Many of these systems operate most efficiently in the pulsed mode and our analysis will be restricted to a fuller understanding of the nature of the transient response of such systems, as perhaps best typified by the CH_3F system pumped with a CO_2 laser. The physical model we use is that of the three level atom or molecule that has been extensively investigated.¹⁵⁻¹⁸

The basic framework of this description of transient behavior in an optically pumped gas, is that of the density matrix equations in the limit in which the time characteristic of the pumping pulse is longer than the relaxation times of the FIR system. In essence, we are examining the transient solution to the equations presented in references 2 and 22. Our formulation of the appropriate equations in no way introduces any new physical component; however, the convenient form in which they are cast permits us to obtain new solutions for the time dependent behavior of optically pumped systems. In that which follows, we shall restrict our attention solely to homogeneously broadened systems.

With regard to earlier solutions obtained for the transient behavior of optically pumped three level systems, we

note that previous authors have postulated phenomenological rate equations that omit certain essential aspects of these systems.^{3,5-9} The features often neglected in these descriptions are associated with non-linear power broadening on both the pump and the FIR fields, and the multiphoton or Raman effects that may be present. These latter transitions are important for the case of off resonant pumping of FIR systems, especially since pump detuning offers the possibility of tunability of FIR frequencies over a fairly wide range of wavelengths.¹⁸⁻²⁰ Solutions to simplified versions of three level rate equations that neglect these effects can yield proper results only in certain limits.

In the following section we briefly develop the equations used, and in section 3 we present our general solution. In section 4, certain asymptotic limits are discussed, and a comparison with some experimental results is made.

2. THREE LEVEL RATE EQUATIONS

2.1 NON-LINEAR RATE EQUATIONS

From the density matrix equations we may properly derive rate equations that retain important power broadening terms and multiphoton effects necessary for a description of the amplification of an optically driven FIR pulse.¹⁴⁻¹⁵ The restriction in this derivation is that the pump pulse be long-

er than the dephasing time of the system, so that we may regard all of the off diagonal terms of the density matrix as synchronized to the instantaneous value of the electromagnetic field.²² Thus, a complete rate equation description is appropriate in all situations in which true coherence effects are absent. A schematic of the physical system we wish to analyze is shown in figure 1, and a description of the detuning parameters is given in figure 2. The actual system we wish to describe is naturally richer in detail than the three level model we employ, and figure 3^{schematically} illustrates some of the true complexities of a typical FIR system. These problems will be discussed below when we calculate physical parameters to make comparison with some experimental results. The consequences of averaging over M and K levels have been quantitatively discussed in a steady state analysis.⁹

On a time scale longer than the true coherence time of the particle internal states, the nature of the interaction between radiation and matter may properly be described by an appropriate line shape function, Q. This contains information on the response of the system to resonant and near resonant radiation and may incorporate important nonlinear broadening effects and multiphoton transitions. In this limit, the density matrix equations, or rather the equations for particle number difference, reduce to the following. We have assumed that the total number of particles in states 1, 2 and 3 is constant.

$$\frac{\partial}{\partial t} \begin{vmatrix} r_{32} \\ r_{13} \end{vmatrix} = - \begin{vmatrix} \tilde{\gamma}_1 & \tilde{\gamma}_2 \\ \tilde{\gamma}_3 & \tilde{\gamma}_4 \end{vmatrix} \begin{vmatrix} r_{32} - r_{32}^0 \\ r_{13} - r_{13}^0 \end{vmatrix} + \begin{vmatrix} 4\beta_{32}^2 & -2\beta_{13}^2 \\ -2\beta_{32}^2 & 4\beta_{13}^2 \end{vmatrix} \text{Im} Q \begin{vmatrix} r_{32} \\ r_{13} \end{vmatrix} \quad (1)$$

Relaxation coefficients $\tilde{\gamma}_{ij}$ are presented in Appendix 1,

$r_{ii} = \rho_{ii} - \rho_{jj}$, and r_{ij}^0 is the equilibrium state difference population. The complex line shape function Q is related to the susceptibility in the following manner.

$$\begin{vmatrix} X(-\omega_s) \\ X(\omega_p) \end{vmatrix} = \frac{1}{\hbar \epsilon_0} \begin{vmatrix} \mu_{32}^2 & 0 \\ 0 & \mu_{13}^2 \end{vmatrix} Q \begin{vmatrix} r_{32} \\ r_{13} \end{vmatrix} \quad (2)$$

The Rabi frequency is given in terms of the electromagnetic field, $\beta_{ij} = \mu_{ij} E_{ij} / \hbar$, and standard notation of reference 1 is employed. The equations for the pump and FIR field amplitudes are

$$\left(\frac{\partial}{\partial t} + C \underline{L} \cdot \underline{\nabla} \right) \begin{vmatrix} \beta_{32}^2 \\ \beta_{13}^2 \end{vmatrix} = - \begin{vmatrix} \gamma_s \beta_{32}^2 \\ \gamma_p \beta_{13}^2 \end{vmatrix} + \begin{vmatrix} -2\omega_s X''(-\omega_s) & 0 \\ 0 & 2\omega_p X''(\omega_p) \end{vmatrix} \begin{vmatrix} \beta_{32}^2 \\ \beta_{13}^2 \end{vmatrix} \quad (3)$$

where γ_s and γ_p are phenomenological loss terms. Spontaneous emission is neglected. It may be incorporated in our solutions as an initial condition on the FIR field intensity. The equations for pump (ϕ_p) and FIR phase (ϕ_s) are then given by the following.

$$\left(\frac{\partial}{\partial t} + C \underline{L} \cdot \underline{\nabla} \right) \begin{vmatrix} \phi_s \\ \phi_p \end{vmatrix} = \begin{vmatrix} \omega_s X'(-\omega_s) \\ -\omega_p X'(\omega_p) \end{vmatrix} \quad (4)$$

where X' and X'' refer, respectively, to the real and imaginary parts of the complex susceptibility. These expressions are algebraically quite involved when presented in

terms of the line shape function (Appendix 1) and equation 2. To simplify these relations, we proceed as follows. Noting the detuning parameters of figure 2, we introduce in Appendix 2 and in figure 4, complex detuning parameters that permit the collisional dephasing frequencies γ_{ij} and the FIR and pump field detuning to be combined as complex values. We now define saturation parameters that are dependent upon detuning as well as upon collisional parameters.

$$I_s^* \equiv \epsilon_0 \hbar^2 C(\alpha_{13} \alpha_{12}) / \mu_{32}^2 \quad (5)$$

$$I_p^* \equiv \epsilon_0 \hbar^2 C(\alpha_{32} \alpha_{12}) / \mu_{13}^2$$

where

$$\alpha_{32} = (\gamma_{32}^2 + \delta_s^2)^{1/2}$$

$$\alpha_{13} = (\gamma_{13}^2 + \delta_p^2)^{1/2} \quad (6)$$

$$\alpha_{12} = (\gamma_{12}^2 + [\delta_s - \delta_p]^2)^{1/2}$$

With these quantities, dimensionless pump and FIR field intensities may be introduced.

$$S \equiv \beta_{32}^2 / \alpha_{13} \alpha_{12} = I_s / I_s^* \quad P \equiv \beta_{13}^2 / \alpha_{32} \alpha_{12} = I_p / I_p^* \quad (7)$$

The complex line shape function Q may now be written in a more compact form as follows.

$$Q = \frac{i}{\Delta} \begin{vmatrix} S + e^{i(\phi - \phi_{32})} & P \\ S & P + e^{i(\phi - \phi_{13})} \end{vmatrix} \begin{vmatrix} \alpha_{32}^{-1} & 0 \\ 0 & \alpha_{13}^{-1} \end{vmatrix} \quad (8)$$

with $\Delta = e^{i\phi} + e^{i\phi_{13}} + e^{i\phi_{32}}$. From figure 4 we see that ϕ is the sum of the three detuning angles, $\phi = \phi_{13} + \theta$, and $\theta = \phi_{32} + \phi_{12}$.

We now are able to write the population difference equations as follows.

$$\frac{\partial}{\partial t} \begin{vmatrix} r_{32} \\ r_{13} \end{vmatrix} = - \begin{vmatrix} \tilde{\gamma}_1 & \tilde{\gamma}_2 \\ \tilde{\gamma}_3 & \tilde{\gamma}_4 \end{vmatrix} \begin{vmatrix} r_{32} - r_{32}^0 \\ r_{13} - r_{13}^0 \end{vmatrix} + \begin{vmatrix} 4\alpha_{13} \alpha_{12} & -2\alpha_{13} \alpha_{12} \\ -2\alpha_{13} \alpha_{32} & 4\alpha_{32} \alpha_{12} \end{vmatrix} \begin{vmatrix} r_{32} \alpha_{32}^{-1} \\ r_{13} \alpha_{32}^{-1} \end{vmatrix} \quad (9)$$

$$\left\{ \text{Re } \frac{1}{\Delta} \begin{vmatrix} S(S + e^{i(\phi - \phi_{32})}) & SP \\ SP & P(P + e^{i(\phi - \phi_{13})}) \end{vmatrix} \begin{vmatrix} r_{32} \alpha_{32}^{-1} \\ r_{13} \alpha_{32}^{-1} \end{vmatrix} \right\}$$

The equations for the FIR and pump intensities and phases are given as

$$\left(\frac{\partial}{\partial t} + C \underline{L} \cdot \underline{\nabla} \right) \begin{vmatrix} S \\ P \end{vmatrix} = - \begin{vmatrix} \gamma_S S \\ \gamma_P P \end{vmatrix} + \frac{1}{\hbar \epsilon_0} \begin{vmatrix} -2\omega_S \mu_{32}^2 & 0 \\ 0 & 2\omega_{12} \mu_{13}^2 \end{vmatrix} \begin{vmatrix} S \\ P \end{vmatrix}$$

$$\left\{ \text{Re } \frac{1}{\Delta} \begin{vmatrix} S(S + e^{i(\phi - \phi_{32})}) & SP \\ SP & P(P + e^{i(\phi - \phi_{13})}) \end{vmatrix} \begin{vmatrix} r_{32} \alpha_{32}^{-1} \\ r_{13} \alpha_{13}^{-1} \end{vmatrix} \right\} \quad (10)$$

$$\left(\frac{\partial}{\partial t} + C \underline{L} \cdot \underline{\nabla} \right) \begin{vmatrix} \phi_S \\ \phi_P \end{vmatrix} = \frac{1}{\hbar \epsilon_0} \begin{vmatrix} \omega_S \mu_{32}^2 & 0 \\ 0 & -\omega_P \mu_{13}^2 \end{vmatrix} \begin{vmatrix} \phi_S \\ \phi_P \end{vmatrix}$$

$$\left\{ \text{Im } \frac{1}{\Delta} \begin{vmatrix} S + e^{i(\phi - \phi_{32})} & P \\ S & P + e^{i(\phi - \phi_{13})} \end{vmatrix} \begin{vmatrix} r_{32} \alpha_{32}^{-1} \\ r_{13} \alpha_{13}^{-1} \end{vmatrix} \right\} \quad (11)$$

Equations 9-11 must be solved with initial conditions specified on the state density differences and the input pump pulse shape. We may take the initial conditions on the FIR field to be either background black body radiation, or a

weak FIR pulse that we wish to amplify. These equations are still formidable and nonlinear, and it will be necessary to consider solutions in the limit in which saturations effects on the FIR pulse may be neglected. This is done in the following section.

2.2 RATE EQUATIONS FOR $S/P \ll 1$

To proceed further toward our goal of a solution with a relative degree of analytical simplicity, it is necessary to make one additional assumption of significance; i.e., that the ratio of the dimensionless FIR field to the dimensionless pump field is small, $S/P \ll 1$, where

$$\frac{S}{P} \equiv \frac{\mu_{32}^2 I_s (\gamma_{32}^2 + \delta_s^2)^{1/2}}{\mu_{13}^2 I_p (\gamma_{13}^2 + \delta_p^2)^{1/2}} \quad (12)$$

Typical values are $\mu_{13} \approx .05$ D, and $\mu_{32} = 1$ D, so that if we set an upper limit of $S/P = .1$, we have then the restriction that $I_s/I_p < 2.5 \times 10^{-4}$. This ratio can be larger for $\delta_p > \delta_s$. Since P decreases as S increases, this description will rigorously hold only for certain lengths of amplifier. For lengths such that saturation in the FIR field plays a role, our results may be only qualitative. With this additional linearization of the FIR field, equations 9-11 become

$$\begin{aligned} \frac{\partial}{\partial \tau} r_{32} = & -\tilde{\gamma}_1 (r_{32} - r_{32}^0) - \tilde{\gamma}_2 (r_{13} - r_{13}^0) \\ & - 2\alpha_{32}\alpha_{12} (r_{13}/\alpha_{13}) P \cos \phi_{13} \end{aligned} \quad (13)$$

$$\begin{aligned} \frac{\partial}{\partial \tau} r_{13} = & -\tilde{\gamma}_3(r_{32} - r_{32}^0) - \tilde{\gamma}_4(r_{13} - r_{13}^0) \\ & + 4\alpha_{32}\alpha_{12}(r_{13}/\alpha_{13}) P \cos \phi_{13} \end{aligned} \quad (14)$$

$$\begin{aligned} \frac{\partial}{\partial z} S = & -\gamma_S S - 2(\mu_{32}^2 \omega_S / \hbar \epsilon_0 C) \left\{ \frac{P \cos \phi_{12} + \cos \phi_{32})(r_{32}/\alpha_{32})}{p^2 + 2 P \cos \theta + 1} \right. \\ & \left. + \left(\frac{p^2 \cos \phi_{13} + P \cos \phi}{p^2 + 2 P \cos \theta + 1} \right) (r_{13}/\alpha_{13}) \right\} S \end{aligned} \quad (15)$$

$$\frac{\partial P}{\partial z} = -\gamma_P P + 2(\mu_{13}^2 \omega_P / \hbar \epsilon_0 C) (r_{13}/\alpha_{13}) P \cos \phi_{13} \quad (16)$$

$$\begin{aligned} \frac{\partial \phi_S}{\partial z} = & (\mu_{32}^2 \omega_S / \hbar \epsilon_0 C) \left\{ \frac{(P \sin \phi_{12} - \sin \phi_{32})}{p^2 + 2 P \cos \theta + 1} (r_{32}/\alpha_{32}) \right. \\ & \left. - \frac{(p^2 \sin \phi_{13} + P \sin \phi)}{p^2 + 2 P \cos \theta + 1} (r_{13}/\alpha_{13}) \right\} \end{aligned} \quad (17)$$

$$\frac{\partial \phi_P}{\partial z} = (\mu_{13}^2 \omega_P / \hbar \epsilon_0 C) (r_{13}/\alpha_{13}) \sin \phi_{13} \quad (18)$$

The equations are valid for large P when saturation and multiphoton effects dominate the solution for S . In the limit of small P , the equation for S takes on the same form as the equation for P . We note that as a consequence of re-

taining power broadening only on the pump line that all equations (except the equation for the FIR field) are independent of S . This is a natural consequence of our neglect of FIR saturation effects. We have also made a standard transformation to a retarded time variable, $\tau = t - z/c$ and have neglected diffusion effects in the equation for state density differences. Equations 13-14 are rate equations that retain nonlinear power broadening effects associated with the pump pulse as well as contributions from multiphoton processes. In that which follows, we shall obtain solutions to equations 13-18 in appropriate limits.

3. SOLUTION OF THE EQUATIONS

Prior to considering a solution for the pump amplitude and phase, which is the key to the solution to our system of equations, we introduce the following complex transfer functions whose use simplifies the FIR and pump equations.

$$A_p(z, \tau) - A_p(0, \tau) \equiv \frac{1}{2} \ln \frac{P(z, \tau)}{P(0, \tau)} - i [\phi_p(z, \tau) - \phi_p(0, \tau)] \quad (19)$$

$$A_s(z, \tau) - A_s(0, \tau) \equiv \frac{1}{2} \ln \frac{S(z, \tau)}{S(0, \tau)} - i [\phi_s(z, \tau) - \phi_s(0, \tau)] \quad (20)$$

With these relations, equations 15-18 may be written more compactly as follows.

$$\left. \frac{\partial A_p}{\partial z} \right|_{\tau} = \frac{-\gamma_p}{2} + \sigma_1 r_{13} e^{-i\phi_{13}} \quad (21)$$

$$\left. \frac{\partial A_s}{\partial z} \right|_{\tau} = \frac{-\gamma_s}{2} - \frac{\sigma_2 r_{32} e^{i\phi_{12}}}{p + e^{i\theta}} - \frac{\sigma_3 r_{13} e^{-i\phi_{13}}}{p + e^{i\theta}} p \quad (22)$$

The three absorption cross sections σ_1 , σ_2 , and σ_3 , are defined in Appendix 2. In order to integrate equation 21, it is necessary to solve for r_{13} . To facilitate this, we assume that all of the T_1 times are equal, and that all of the T_2 times are equal. Thus, as can be seen from Appendix 1, $\gamma_{11} = \gamma_{22} = \gamma_{33} = 1/T_1$, $\tilde{\gamma}_2 = \tilde{\gamma}_4 = 0$, $\tilde{\gamma} = \tilde{\gamma}_1 = \tilde{\gamma}_3 = 1/T_1$. Further

$$\gamma \equiv -4(\alpha_{32}\alpha_{12} \cos \phi_{12}/\alpha_{13}) = 4/T_2 \quad (23)$$

where T_2 is the collisional dephasing time.

At this point, it is convenient to define ϕ , a pump pulse area that retains the effects of collisional deactivation.

$$\phi \equiv \int_{-\infty}^{\tau} e^{-\tilde{\gamma}(\tau - t)} P(z, t) \gamma dt \quad (24)$$

If we now, in addition, define a density function ρ such that

$$\rho \equiv \int_{-\infty}^{\tau} e^{-\tilde{\gamma}(\tau - t)} r_{13} P \gamma dt = \int_{\phi_0}^{\phi} r_{13} d\phi \quad (25)$$

then equations 13 and 14 take on extremely simple forms.

$$\frac{\partial r_{32}}{\partial \tau} + \tilde{\gamma} r_{32} = \tilde{\gamma} r_{32}^0 + \frac{\gamma}{2} r_{13}^P \quad (26)$$

$$\frac{\partial r_{13}}{\partial \tau} + \tilde{\gamma} r_{13} = \tilde{\gamma} r_{13}^0 - \gamma r_{13}^P \quad (27)$$

These may be formally integrated to yield,

$$r_{32} = r_{32}^0 + \rho/2 \quad (28)$$

$$r_{13} = r_{13}^0 - \rho \quad (29)$$

When these results are substituted into equations 21 and 22, we may integrate to obtain the following expressions.

$$A_p(z, \tau) - A_p(0, \tau) = \sigma_1 r_{13}^0 e^{-i\phi_{13}} z - \sigma_1 \int_{\phi_0}^{\phi} \rho e^{i\phi_{13}} \left(\frac{\partial z}{\partial \phi} \right)_{\tau} d\phi \quad (30)$$

$$A_s(z, \tau) - A_s(0, \tau) = \frac{-\gamma_s}{2} z - \sigma_2 \int_{\phi_0}^{\phi} \frac{[r_{32}^0 + \frac{\rho}{2}] e^{i\phi_{12}} \left(\frac{\partial z}{\partial \phi} \right)_{\tau}}{p + e^{i\theta}} d\phi \quad (31)$$

$$- \sigma_3 \int_{\phi_0}^{\phi} [r_{13}^0 - \rho] \frac{p e^{-i\phi_{13}} \left(\frac{\partial z}{\partial \phi} \right)_{\tau}}{p + e^{i\theta}} d\phi$$

Equation 30 and 31 are formal results. Not only must we obtain ρ and $(\partial \phi / \partial z)_{\tau}$ as functions of ϕ to perform the integration, we must somehow obtain ϕ as a function of the pump pulse P , or, equivalently, $P = P(\phi)$. In order to proceed further, we make the additional assumption that the cavity loss parameter for the pump pulse, γ_p , may be neglected. Multiplying equation 16 by $e^{-\tilde{\gamma}(\tau-t)}$ and integrating, we find that

$$\left(\frac{\partial \phi}{\partial z} \right)_{\tau} = -\sigma \rho \quad (32)$$

where $\sigma = -2 \sigma_1 \cos \phi_{13}$. This permits the integration of equation 30.

$$p(z, \tau) - A_p(0, \tau) = z r_{13}^0 \sigma_1 e^{-i\phi_{13}} + (\sigma_1 / \sigma) e^{-i\phi_{13}} (\phi - \phi_0) \quad (33)$$

We now may separate the real and imaginary parts of the above solution to obtain the pump amplitude and phase as functions of position and ϕ .

$$\ln \frac{P(z, \tau)}{P(0, \tau)} = - \frac{2\mu_{13}^2 \omega_p r_{13}^0 z \cos^2 \phi_{13}}{\hbar \epsilon_0 C \gamma_{13}} + (\phi_0 - \phi) \quad (34)$$

(35)

$$\phi_p(z, \tau) - \phi_p(0, \tau) = - (\mu_{13}^2 \omega_p r_{13}^0 z / \hbar \epsilon_0 C \gamma_{13}) \sin \phi_{13} \cos \phi_{13} + \frac{1}{2} (\phi_0 - \phi) \tan \phi_{13}$$

Equations 34 and 35 represent a solution to the behavior of the pump pulse if $\phi = \phi(P)$ is known. As previously indicated, we will examine this relationship in the limit in which the time characterizing the pump pulse half width is longer than either the T_1 or T_2 relaxation times. This approximation corresponds to many optically pumped FIR laser systems pumped by a CO_2 TEA laser.

For the case in which $\tau_p / T_1 \gg 1$, we may take the limit in which $\tilde{\gamma}$ becomes large. Thus, the exponential in equation 24 may be replaced by a δ function, and

$$\phi = \frac{\gamma}{\tilde{\gamma}} P = \frac{4T_1}{T_2} P \quad (36)$$

From equation 34, we obtain the following result.

$$P/P_0 = e^{-\sigma r_{13}^0 z} \exp \left[+ \frac{\gamma}{\tilde{\gamma}} (P_0 - P) \right] \quad (37)$$

This simple equation defines the spatial and temporal variation of the pump pulse as a function of the cross section σ , the initial state densities r_{13}^0 , and the ratio of relaxation times $4T_1/T_2$, or equivalently, $\gamma / \tilde{\gamma}$. The dependence of P upon the retarded time appears only through $P_0(0, \tau)$, the initial condition on the pump pulse. Equation 37 will be compared with some experimental results below. The solution for $P(\phi)$ to be substituted into our expression for the FIR field is given by equation 36. The remaining quantity to be determined in order to be able to integrate equation 31 is $\rho(\phi)$. We note the singular nature of the limit in which $\tilde{\gamma}$ becomes large by writing equation 27 in the following manner.

$$\left(\frac{\partial r_{13}}{\partial \phi} \right)_z \left(\frac{\partial \phi}{\partial \tau} \right)_z = -\tilde{\gamma} (r_{13} - r_{13}^0) - \gamma r_{13} P \quad (38)$$

Differentiating equation 24 with respect to τ , and substituting into the above relation, we find

$$[P\gamma - \tilde{\gamma}\phi] \left(\frac{\partial r_{13}}{\partial \phi} \right)_\tau = -\tilde{\gamma}(r_{13} - r_{13}^0) - \gamma r_{13} P \quad (39)$$

In the limit of large $\tilde{\gamma}$, the left hand side of equation 39 vanishes, and we obtain an algebraic dependence of ρ upon ϕ . This merely indicates the limit in which, for the pumping pulse longer than the relaxation times of the system, the state densities respond instantaneously to changes in the value of the pump field. Thus,

$$r_{13} = r_{13}^0 / (1 + \phi) \quad \rho = \phi r_{13}^0 / (1 + \phi) \quad (40)$$

We now have the relations for $P(\phi)$, $\rho(\phi)$, and $r_{13}(\phi)$ to be substituted into equation 31. The integrations may now be explicitly carried out.

$$A_s - A_{s0} = -\frac{\gamma_s z}{2} - \frac{\mu_{32}^2 \omega_s}{2\mu_{13}^2 \omega_p \cos \phi_{13}} \left[\frac{\alpha_{13}}{\alpha_{32}} \frac{e^{i\phi_{12}}}{2} \frac{\gamma}{\tilde{\gamma}} + e^{-i\phi_{13}} \right] \ln \frac{P + e^{i\theta}}{P_0 + e^{i\theta}} \quad (41)$$

Resolving equation 41 into real and imaginary components, we obtain solutions for the FIR intensity and phase.

$$\ln \frac{S(z, \tau)}{S(0, \tau)} = -\gamma_s z + \frac{\mu_{32}^2 \omega_s}{\mu_{13}^2 \omega_p} (a + 1) \frac{1}{2} \ln \left(\frac{x_0^2 + 1}{x^2 + 1} \right) \quad (42)$$

$$+ \frac{\mu_{32}^2 \omega_s}{\mu_{13}^2 \omega_p} (a \tan \phi_{12} - \tan \phi_{13}) \tan^{-1} \left(\frac{x_0 - x}{1 + x_0 x} \right)$$

$$\phi_s(z, \tau) - \phi_s(0, \tau) = \frac{1}{2} \frac{\mu_{32}^2 \omega_s}{\mu_{13}^2 \omega_p} (a + 1) \tan^{-1} \left(\frac{x_0 - x}{1 + x_0 x} \right) \quad (43)$$

$$- \frac{1}{4} \frac{\mu_{32}^2 \omega_s}{\mu_{13}^2 \omega_p} (a \tan \phi_{12} - \tan \phi_{13}) \ln \left(\frac{x_0^2 + 1}{x^2 + 1} \right)$$

where

$$x = \frac{P + \cos \theta}{\sin \theta} \quad (44)$$

$$a = \frac{\gamma}{2\tilde{\gamma}} \frac{\alpha_{13}}{\alpha_{32}} \frac{\cos \phi_{12}}{\cos \phi_{13}}$$

These solutions have required the following assumptions.

1. The pumping pulse τ_p is longer than either of the relaxation times T_1 or T_2 .
2. The cavity losses on the pump pulse, γ_p , have been neglected.
3. The total population of the three states has been taken as constant.
4. We have as-

sumed that saturation effects on the FIR field may be neglected, i.e. $S/P \ll 1$. 5. Only homogeneous broadening is considered. 6. We have also neglected r_{32}^0/r_{13}^0 .

These equations represent the response of a three level system to an arbitrary pump pulse, subject to the assumptions just noted. An extremely simple form results for those conditions in which detuning on both fields is absent. This will be discussed in the following section.

4. ASYMPTOTIC LIMITS AND COMPARISON WITH EXPERIMENT

4.1 ASYMPTOTIC LIMITS

We first consider the behavior of the pump pulse in terms of a more convenient parameter, the absorption, which is defined as follows.

$$A(z, \tau) = 1 - P(z, \tau)/P_0 \quad (45)$$

and a dimensionless absorption length,

$$\beta = \sigma r_{13}^0 z \quad (46)$$

We may now write equation 37 as follows.

$$\beta = \phi_0 A - \ln(1 - A) \quad (47)$$

Thus, the absorption of the pumping pulse may be expressed as $A(\beta, \phi_0)$. This dependence is shown in figure 5. It is to be noted that the absorption is only a function of the

initial value of the pump pulse, which is in itself a function of the retarded time, and the dimensionless absorption length β . We will see below that this simple result compares well with experiment. Consider now the FIR field intensity as given by equation 42. The algebraic complexity of this result indicates that it is perhaps of value to examine the case in which there is zero detuning i.e., $\delta_s = \delta_D = 0$. Thus, $\phi_{12} = \phi_{23} = \phi_{12} = \pi$, $\theta = 2\pi$, and our result for the solution of the FIR field intensity is given as follows.

$$\frac{I_s}{I_{s0}} = \frac{S}{S_0} = e^{-\gamma_s z} \left[\frac{P_0 + 1}{P + 1} \right]^\xi \quad (48)$$

where

$$\xi = \frac{\mu_{32}^2 \omega_s}{\mu^2 \omega_p} \left(\frac{\gamma}{2\gamma} + 1 \right) \quad (49)$$

We note from equation 48 that for $P_0 \gg 1$, if P becomes small, that $I_s/I_{s0} \sim (P_0(\tau))^\xi$. A typical value of ξ would be of the order of ten, so that for these conditions, there is a considerable steepening or compression of the FIR pulse.

We may also define a reduced FIR gain coefficient in terms of pump absorption A , P_0 , the initial pump pulse, and ξ .

$$\left[\frac{I_s}{I_{s0}} e^{\gamma_s z} \right]^{1/\xi} \equiv n \equiv \frac{1}{1 - \left(\frac{P_0}{P_0 + 1} \right) A} \quad (50)$$

and this is presented in figure 6. Thus, we have an extremely simple result for the growth of the FIR intensity that parametrically depends upon the amount of pump power absorbed. If we then consider the situation of strong pumping, $P \gg 1$, then we find

$$n = \frac{1}{1 - A} \quad (51)$$

For $A \approx 1$, our results are invalid, since at this point the assumption of weak saturation on the FIR transition will have been violated.

We note, however, that not only β but P_0 (or ϕ_0) and A are dependent upon pressure. For the case of the homogeneously broadened pumping pulse considered in the present analysis, σ_{13} , the absorption coefficient of equation 46 is proportional to pressure. It is therefore convenient to define a new parameter ϵ that is independent of pressure, but proportional to pump input power and cavity length. With $\epsilon \equiv \beta \phi_0$ we may write equation 47 as $A = A(\epsilon, \beta)$, and curves of constant ϵ correspond to curves of fixed cavity length, and a fixed pumping power. The variation of pump absorption with pressure occurs only through the pressure dependence of the dimensionless absorption length β . See figure 7. We may also use equations 47 and 50 to obtain a relation for $n = n(A, \epsilon)$.

$$n = 1 + \frac{A}{1 - A + \frac{2}{\epsilon} \{ \sqrt{\ln(1 - A)^2 + 4\epsilon A} - \ln(1 - A) \}} \quad (52)$$

This relation is presented in figure 8. Curves of constant ϵ are independent of pressure, and the reduced FIR gain coefficient is conveniently given in terms of ϵ and the absorption A . We see from figure 8 that at low values of ϵ , corresponding to low input power or short cavities, there is a broad maximum in the FIR gain coefficient with respect to absorption. This maximum shifts toward higher values of the absorption as cavity length or input power are increased. We should also note, that for very high values of absorption, our results can not be expected to provide quantitative agreement with experiment, since the restriction that $S/P \ll 1$ will have been violated.

4.2 COMPARISON WITH EXPERIMENT

In order to obtain a comparison with experiment it is necessary to obtain values for r_{13}^0 and ϕ that correspond to a specific optically pumped system. We consider the 496 μm FIR CH_3F laser pumped by the $\text{P}(20)$ 9.55 μm CO_2 line.²⁴⁻²⁹ Saturation absorption of this line has been measured by Temkin, et. al.³⁰ and the FIR gain by Brown, et. al.³¹ The CO_2 pumping pulse in the experiments have a typical length of 100 nanoseconds (FWHM) and the relaxation times are approximately 10 nanoseconds/p, where p is the pressure in torr. These conditions are consistent with the assumptions made in obtaining our solutions. We shall now assume that $T_1 = T_2 = 1/\gamma_{11} = 1/\gamma_{22}$. To calculate the value of

the absorption cross section for the pumping pulse, we note that,

$$\sigma = \frac{2\mu_{13}^2 \omega_p \gamma_{13}}{\epsilon_0 \hbar C (\gamma_{13}^2 + \delta_p^2)} \quad (53)$$

where γ_{13} characterizes the homogeneous linewidth of the pumping pulse $\Delta \nu_{\text{CO}_2} \sim 2 \text{ GHz}$, and

$$\gamma_{13} = \sqrt{\frac{\pi}{\ln 2}} \Delta \nu_{\text{CO}_2} \approx 4.26 \times 10^9 \text{ sec.}^{-1} \quad (54)$$

The remaining quantity to be determined in order to obtain an absorption cross section, is an average dipole moment for CH_3F for the transition of interest. Since our simplified model encompasses only three levels, it is necessary to sum over the M and K quantum levels in an appropriate fashion. This calculation is outlined in Appendix III, and we obtain a value of $\langle \mu_{13} \rangle = .00213 \text{ D}$, to be substituted into equation 53. With this result, the average of the cross section is given as $\sigma = 1.67 \times 10^{-19} \text{ cm}^2$. The initial state density of those particles interacting with the pump pulse is calculated to be $r_{13}^0 = 3.22 \times 10^{16} \text{ p cm}^{-3}$, where again, p is the pressure in torr. Thus the total absorption cross section is given by $\sigma r_{13}^0 = 5.38 \times 10^{-3} \text{ p cm}^{-1}$. These values for the state density are taken at a temperature of 300 K. The dimensionless intensity, or initial pulse area is given as follows,

$$\phi_0 = \sigma \left(\frac{1}{\gamma_{11}} + \frac{1}{\gamma_{33}} \right) \frac{I_0}{\hbar \nu_p} \quad (55)$$

However, the total decay rates of level 1 and level 3 are the summations of contributions from all active sublevels weighted by the Boltzmann factor, thus, we must sum

$$\gamma_{11} = \sum_K \sum_M f_{JKM} \gamma_{JKM} = f/T_1$$

where f_{JKM} is the statistical weight of the sublevels. We calculate an effective Boltzmann constant $f = .1118$. Thus for $T_1 = 10 \times 10^{-9}$ sec/p, $\gamma_{11} = 1.118 \times 10^7$ sec/p and for the parameters calculated above, we find that $\phi_0 = 1.44 \times 10^{-6} I_{0/p}$, where I_0 is the intensity in watts/cm², and p the pressure in torr. From equation 49, with $\mu_{13} = .05$ D, $\mu_{32} = 1$ D, we find the value of ξ to be 24.

With a pump pulse of .5 J and a FWHM of 100 nanoseconds, we see, using equations 46 and 47, a comparison of experiment and theory. This is presented in figures 9 and 10 along with the numerical results calculated by Pichamuthu and Sinha.⁶ Thus, equation 47 seems an adequate prediction of the behavior of optical absorption by a pumping pulse, and only two parameters need be obtained to produce a satisfactory agreement with experiment. The curves in figures 9 and 10 were obtained by utilizing the fact that σ is proportional to pressure, and that ϕ_0 is inversely proportional to pressure. From equation 49, we calculate the value of ξ to be 24. Further, once having obtained the value of the absorption and $\beta\phi_0$, we may compare the reduced gain coefficient, equation 52, with the experiments of Brown, et. al.³¹. A slightly different value of the absorption coeffi-

cient was used for this comparison, $\beta r S^{0.13} = .003 \text{ p cm}^{-1}$ and $\phi_0 = 3.5 p_0$. It is seen that agreement between the theory presented here and the experimental results for FIR gain is satisfactory. The close correlation with the experiment for $A \sim 1$ perhaps indicates that for these low values of the FIR reduced gain coefficient, that saturation effects do not play a dominant role.

5. SUMMARY

In the limit in which the pumping pulse is long compared with the longitudinal and transverse relaxation times of an optically pumped system, we have obtained time dependent solutions for the absorption of an optical pumping pulse and the subsequent generation of an FIR pulse. The solution for the absorption is particularly simple, and seems to compare well with several sets of experiments. The absorption depends only upon an effective absorption coefficient, and, in a highly nonlinear manner, upon the initial shape of the pumping pulse. Since we have neglected saturation effects on the FIR wavelength, the pumping pulse is not influenced by the presence of the FIR pulse. This can only be true for certain values of the absorption or certain lengths of amplifier. The solution for the pumping pulse permits a solution of the response of the FIR system, and by recasting the equations for FIR field intensity, phase, and population density differences in a particularly convenient form, it

has been possible to solve this system of equations parametrically in terms of the pumping pulse solution. The FIR field solution depends upon a host of molecular parameters, upon the behavior of the input pumping pulse, and upon the pumping absorption coefficient. It also depends in a complicated manner, upon the detuning parameters of the system for both the pump and the FIR signal fields. In the limit in which detuning may be neglected, particularly simple results may be obtained for this complicated system, and these results have been shown to compare favorably with some experiments. The simple analytical form shows the specific role that the initial pumping pulse can have in the shaping of the FIR field intensity, and illustrates how this dependence can lead to pulse narrowing effects as the FIR field intensity grows.

6. ACKNOWLEDGEMENTS

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REFERENCES

1. R. J. Temkin, Theory of Optically Pumped Submillimeter Lasers, IEEE J. Quant Electron., Vol. QE-13, No. 6, p. 450, 1977.
2. R. L. Panock and R. J. Temkin, Interaction of Two Laser Fields with a Three-level Molecular System, IEEE J. Quant. Electron., Vol. QE-13, p. 425, 1977.
3. J. O. Henningsen and H. G. Jensen, The Optically Pumped Far-Infrared Laser: Rate Equations and Diagnostic Experiments, IEEE J. Quant. Electron., Vol. QE-11, No. 6, p. 248, 1975.
4. J. R. Tucker, Absorption Saturation and gain in Pulsed CH_3F Lasers, Optics Commun., Vol. 16, p. 204, 1976.
5. R. J. Temkin and D. R. Cohn, Rate Equations for an Optically Pumped, Far-Infrared Laser, Optics Commun., Vol. 16, No. 2, 1976.
6. J. P. Pichamuthu and U. N. Sinha, Rate Analysis of the Pulsed Colinearly Pumped Far-Infrared Amplifier, IEEE J. Quant. Electron, Vol. QE-15, p. 501, 1979.
7. J. P. Pichamuthu and U. N. Sinha, Saturable Absorption of CO_2 -Laser Radiation by CH_3F , Optics Commun., Vol. 24, p. 195, 1978.
8. T. Y. Chang and T. J. Bridges, Opt. Comm., Vol. 1, p. 423, 1970.
9. H. J. A. Bluysen, R. E. McIntosh, A. F. von Etteger, and P. Wyder, Pulsed Operation of an Optically Pumped Far-Infrared Molecular Laser, IEEE J. Quant. Electron., Vol. QE-11, p. 34, 1975.
10. Zbigniew Drozdowics, R. J. Temkin, and B. Lax, Laser Pumped Molecular Lasers-Part I: Theory, IEEE J. Quant. Electron., Vol. QE-15, No. 3, 1979.
11. Z. Drozdowicz, R. J. Temkin, and B. Lax, Laser Pumped Molecular Lasers, Part II: Submillimeter Laser Experiments, IEEE J. Quant. Electron., Vol. QE-15, p. 865, 1979.
12. J. Heppner, C. O. Weiss, U. Huebner, and G. Schinn, Gain in CW Laser Pumped FIR Laser Gases, IEEE J. Quant. Electron., Vol. QE-16, p. 392, 1980.
13. G. D. Willenberg, U. Huebner, and J. Heppner, Far-Infrared CW Lasing in NH_3 , Optics Comm., Vol, 33, p. 193, 1980.
14. J. Heppner and U. Huebner, Gain saturation of CW Laser Pumped FIR Laser Gases, IEEE J. Quant. Electron., QE-16, Oct. 1980.
15. T. A. DeTemple, Pulsed Optically Pumped Far Infrared Lasers, in Infrared and Submillimeter Waves, edited by Kenneth J. Button, Academic Press 1979.

16. T. Yajima, Three-level Maser Action in a Gas, 1, Theory of Multiple Quantum Transitions and Doppler Effects in the Three Level Gas Maser, J. Phys. Soc. Japan, Vol. 16, pp. 1594-1604, Aug. 1961.
17. A. Javan, Theory of a Three Level Maser, Phys. Rev., Vol. 107, pp. 1597-1589, Sept. 15, 1957.
18. I. M. Beterov and V. P. Chebotaev, Three Level Gas Systems and Their Interaction with Radiation, in Progress in Quantum Electronics, Vol. 3, Part I, J. H. Sanders and S. Stenholm, Ed., Oxford, Pergamon Press, 1974.
19. S. J. Petuchowski, A. T. Rosenberger, and Thomas A. DeTemple, Stimulated Raman Emission in Infrared Excited Gases, IEEE J. Quant. Electron., Q.E.-13, No. 6, 1977.
20. R. Frey, F. Pradere, and J. Ducuing, Tunable Far-Infrared Raman Generation, Optics Commun., Vol. 23, p. 65, Oct. 1977.
21. H. R. Fetterman, H. R. Schlossberg, and Jerry Waldman, Submillimeter Waves Optically Pumped Off Resonance, Optics Commun., Vol. 6, No. 2, p. 165, Oct. 1972.
22. T. Tsai and T. F. Morse, Rate Equations for a Three-Level System, IEEE J. Quant. Electron., Vol. QE-15, p. 1334, 1979.
23. T. J. McIlrath and J. L. Carlsten, Populating Excited States of Incoherent Atoms using Coherent Light, Phys. Rev. A, Vol. 6, No. 3, p. 1091, Sept. 1972.
24. T. A. DeTemple and E. J. Danielewicz, Continuous Wave CH_3F Waveguide Laser at 496 μm : Theory and Experiment, IEEE J. Quant. Electron., Vol. QE-12, p. 40 1976.
25. T. K. Plant and T. A. DeTemple, Configurations for High-Power Pulsed CH_3F 496- μm Lasers, J. Appl. Phys., Vol. 47, p. 3042, 1976.
26. A. Semet and N. E. Luhman, Jr., High Power Narrow Line Pulsed 496 μm Laser, Appl. Phys. Lett., Vol. 28, p. 659, 1976.
27. D. E. Evans, L. E. Sharp, B. W. James, and W. A. Peebles, Far-Infrared Superradiant Laser Action in Methyl Fluoride, Appl. Phys. Lett., Vol. 26, p. 630, 1975.
28. L. E. Sharp, W. A. Peebles, B. W. James, and D. E. Evans, 10 KW Cavity Operation of a Submillimeter CH_3F Laser, Optics Commun., Vol. 14, p. 215, 1975.
29. Gerhard A. Koepf and Kenneth Smith, The CW-496 μm Methylfluoride Laser: Review and Theoretical Predictions, IEEE J. Quant. Electron., Vol. QE-14, No. 5, May 1978.
30. R. J. Temkin, D. R. Cohn, Z. Drozdowicz, Pumping and Emission Characteristics of a 4 kw, Submillimeter CH_3F Laser, Optics Commun., Vol. 14, p. 314, 1975.

31. F. Brown, P. D. Hislop and S. R. Kronheim, Characteristics of a Linearly Pumped Laser Oscillator-Amplifier at 496 μm , Appl. Phys. Lett., Vol. 654, 1976.
32. S. M. Freund, G. Duxbury, M. Romheld, J. T. Tiedje, and T. Oka, Laser Stark Spectroscopy in the 10 μm Region: The Bands of CH_3F , J. Molec. Spect., Vol. 52, p. 38, 1974.
33. C. H. Townes and A. L. Schawlow, Microwave Spectroscopy, New York, McGraw-Hill, 1955, Chap. 3.

Appendix I

The $\tilde{\gamma}_i$ are given as follows.

$$\tilde{\gamma}_1 = \frac{\gamma_{22}(\gamma_{11} + 2\gamma_{33})}{\gamma_{11} + \gamma_{22} + \gamma_{33}}$$

$$\tilde{\gamma}_2 = \frac{\gamma_{11}(\gamma_{22} - \gamma_{33})}{\gamma_{11} + \gamma_{22} + \gamma_{33}}$$

$$\tilde{\gamma}_3 = \frac{\gamma_{22}(\gamma_{11} - \gamma_{33})}{\gamma_{11} + \gamma_{22} + \gamma_{33}}$$

$$\tilde{\gamma}_4 = \frac{\gamma_{11}(\gamma_{22} + 2\gamma_{33})}{\gamma_{11} + \gamma_{22} + \gamma_{33}}$$

where γ_{ii} is the total relaxation out of state ii.

The real and the imaginary parts of the line shape function Q, are given, in the notation of Ref. 1, as follows.

$$\begin{aligned} \text{Re}Q = & \begin{vmatrix} \frac{\delta_s}{\gamma_{32}^2 + \delta_s^2} & 0 \\ 0 & \frac{-\delta_p}{\gamma_{13}^2 + \delta_p^2} \end{vmatrix} + \frac{1}{D_1^2 + D_2^2} \begin{vmatrix} \frac{(D_2^{\alpha_1} + D_1^{\alpha_2})}{\gamma_{32}^2 + \delta_s^2} \beta_{13} \beta_{31} D_2^{\beta_{31}} \beta_{23} \\ D_2^{\beta_{32}} \beta_{23} \frac{(D_2^{\alpha_1} - D_1^{\alpha_2})}{\gamma_{13}^2 + \delta_p^2} \beta_{32} \beta_{23} \end{vmatrix} \\ \text{Im}Q = & \begin{vmatrix} \frac{-\gamma_{32}}{\gamma_{32}^2 + \delta_s^2} & 0 \\ 0 & \frac{-\gamma_{13}}{\gamma_{13}^2 + \delta_p^2} \end{vmatrix} + \frac{1}{D_1^2 + D_2^2} \begin{vmatrix} \frac{(D_1^{\alpha_1} - D_2^{\alpha_2})}{\gamma_{32}^2 + \delta_s^2} \beta_{13} \beta_{31} D_1^{\beta_{13}} \beta_{31} \\ D_1^{\beta_{32}} \beta_{23} \frac{(D_1^{\alpha_1} + D_2^{\alpha_2})}{\gamma_{13}^2 + \delta_p^2} \beta_{32} \beta_{23} \end{vmatrix} \end{aligned}$$

with

$$D_1 = (\gamma_{13}\delta_s - \gamma_{32}\delta_p)(\delta_s - \delta_p) - \gamma_{12}(\gamma_{32}\gamma_{13} + \delta_s\delta_p) - \gamma_{32}\beta_{32}\beta_{23} - \gamma_{13}\beta_{13}\beta_{31}$$

$$D_2 = (\delta_s \delta_p + \gamma_{13} \gamma_{32}) (\delta_s - \delta_p) + \gamma_{12} (\gamma_{13} \delta_s - \gamma_{32} \delta_p) + \delta_s \beta_{32} \beta_{23} - \delta_p \beta_{13} \beta_{31}$$

$$\alpha_1 = \delta_s \delta_p - \gamma_{32} \gamma_{13}$$

$$\alpha_2 = \gamma_{13} \delta_s + \gamma_{32} \delta_p$$

Appendix II

A phasor diagram, (see Fig. 4), permits us to present the collisional dephasing γ_{ij} and the pump and field detuning as complex values.

$$\Delta_{ij} \equiv \alpha_{ij} e^{i\phi_{ij}}$$

$$\alpha_{32} = (\gamma_{32}^2 + \delta_s^2)^{1/2}$$

$$\alpha_{13} = (\gamma_{13}^2 + \delta_p^2)^{1/2}$$

$$\alpha_{12} = ([\delta_s - \delta_p]^2 + \gamma_{12}^2)^{1/2}$$

Detuning angles are given by

$$\phi_{32} = \pi - \tan^{-1}(\delta_s/\gamma_{32})$$

$$\phi_{13} = \pi + \tan^{-1}(\delta_p/\gamma_{13})$$

$$\phi_{12} = \pi - \tan^{-1}[(\delta_s - \delta_p)/\gamma_{12}]$$

$$\phi \equiv \phi_{12} + \phi_{13} + \phi_{32}, \quad \theta \equiv \phi_{32} + \phi_{12}$$

Absorption Cross Sections

$$\sigma_1 = \frac{\mu_{13}^2 w_p}{\hbar \epsilon_0 c \alpha_{13}},$$

$$\sigma_2 = \frac{\mu_{32}^2 w_s}{\hbar \epsilon_0 c \alpha_{32}},$$

$$\sigma_3 = \frac{\mu_{32}^2 w_s}{\hbar \epsilon_0 c \alpha_{13}}$$

Appendix III

To calculate a value of μ_{13} that we use in our three level model, we must appropriately average over all of the M and K sublevels of the transitions that are capable of interacting with the pumping radiation. The single narrow band absorption coefficient of each JKM sublevel is given as

$$\alpha_{JKM} = \left(\frac{4\pi}{\epsilon_0 hc} \right)^2 \mu_{JKM}^2 \left[\frac{\nu(\Delta\nu_h/2)}{(\Delta\nu_h/2)^2 + (\nu - Q(J,K))^2} \right] (r_{13}^0)_{JKM}$$

where $Q(J,K)$ is the frequency of the Q branch transition ($\nu_3 = 1, J = 12, K \rightarrow \nu_3 = 0, J = 12, k$), and the individual dipole of each JKM sublevel is given by

$$\mu_{JKM}^2 = \frac{\mu_K^2 \mu_M^2}{J^2 (J+1)^2}$$

Freund, et. al.³² has determined μ in the above equation to be 0.04686 D.

The difference density $(r_{13}^0)_{JKM}$ is related to the total vibrational state density difference through the Boltzmann factor,

$$\frac{(r_{13}^0)_{JKM}}{(r_{13}^0)} = f_{JKM}$$

and, using the values for CH_3F from reference 33, we calculate at 300 K that $f = 0.1118$.

Since each absorption coefficient α_{JKM} is frequency dependent through the Lorentzian lineshape, the total absorption will depend on the pump intensity at that frequency. An average absorption coefficient $(\sigma r_{13}^0)_{av}$ will be given by

$$(\sigma r_{13}^0)_{av} = \sum_K \sum_M \int_{-\infty}^{\infty} \alpha_{JKM} \cdot \frac{I_p(\nu)}{I_0} d\nu$$

We assume the CO_2 TEA laser has a Gaussian spectral composition centered at $P(20)$, with a bandwidth (FWHM) of $\Delta\nu_{\text{CO}_2} = 2\text{GHz}$.

$$\frac{I_p(\nu)}{I_0} = \left(\frac{1}{\Delta\nu_{\text{CO}_2}} \right) \sqrt{\frac{4 \ln 2}{\pi}} \left[\exp - \left(\frac{2\sqrt{\ln 2}}{\Delta\nu_{\text{CO}_2}} (\nu - P(20)) \right)^2 \right]$$

where I_0 is the total intensity integrated over the spectral distribution of the exciting CO_2 laser beam.

The homogeneous linewidth of each individual K sublevel is approximately $\Delta\nu_h = 40 \text{ p MHz}$, which is small compared with the carbon dioxide linewidth of 2 gigahertz. Therefore, each individual K sublevel may be considered to be excited by a broadband pumping source. In the broadband pumping limit considered here, the Lorentzian appearing in the single narrow band absorption coefficient behaves as a δ function in the above integral over the Gaussian frequency distribution of the pumping pulse intensity. The indicated integration may then be carried out. This permits the identification of the equivalent level constants that must appear in equation 53.

$$\mu_{13}^2 = \frac{\mu^2}{3} \sum_{K=12}^{12} f_{JKM} \frac{(2J+1)}{J(J+1)} K^2 \exp - \left[\left(\frac{\sqrt{4 \ln 2}}{\Delta\nu_{\text{CO}_2}} (Q(J,K) - P(20)) \right)^2 \right]$$

When the values for f_{JKM} , $J = 12$, $\mu = .0469 \text{ D}$ are substituted into the above expression, we find that $\mu_{13} = 0.00213 \text{ D}$.

— PUMP PULSE

- - - FIR PULSE

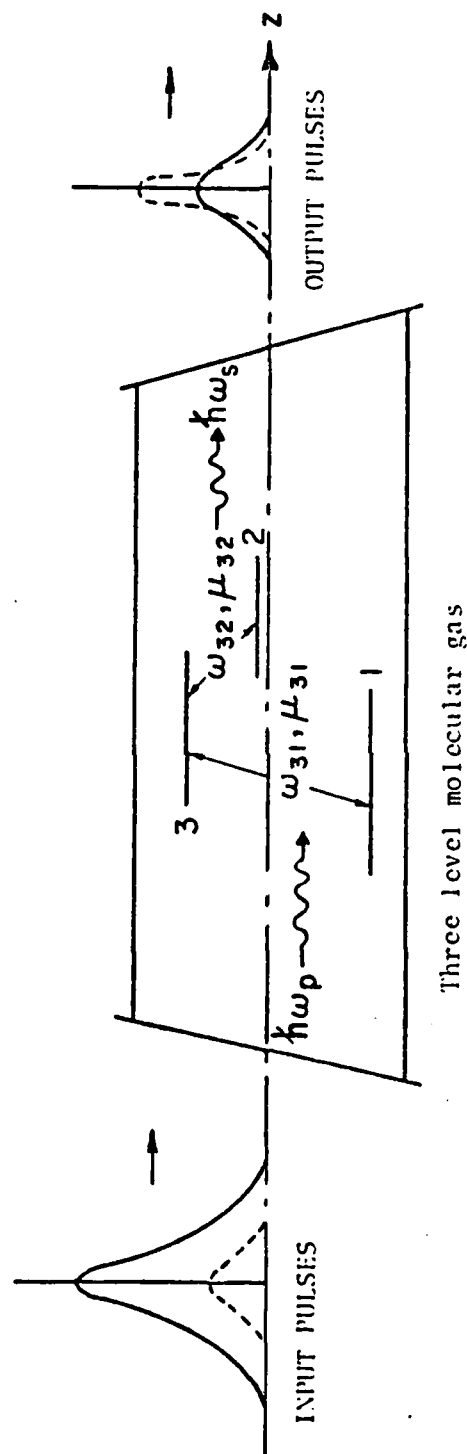


Fig. 1 Pulse Propagation in a Three Level Medium

$$\theta_{13} = \pi |u_{13}| E_1 / \hbar$$

$$\delta_p = \omega_p - \omega_{31}$$

$$\theta_{32} = \pi |u_{32}| E_s / \hbar$$

$$\delta_s = \omega_s - \omega_{32}$$

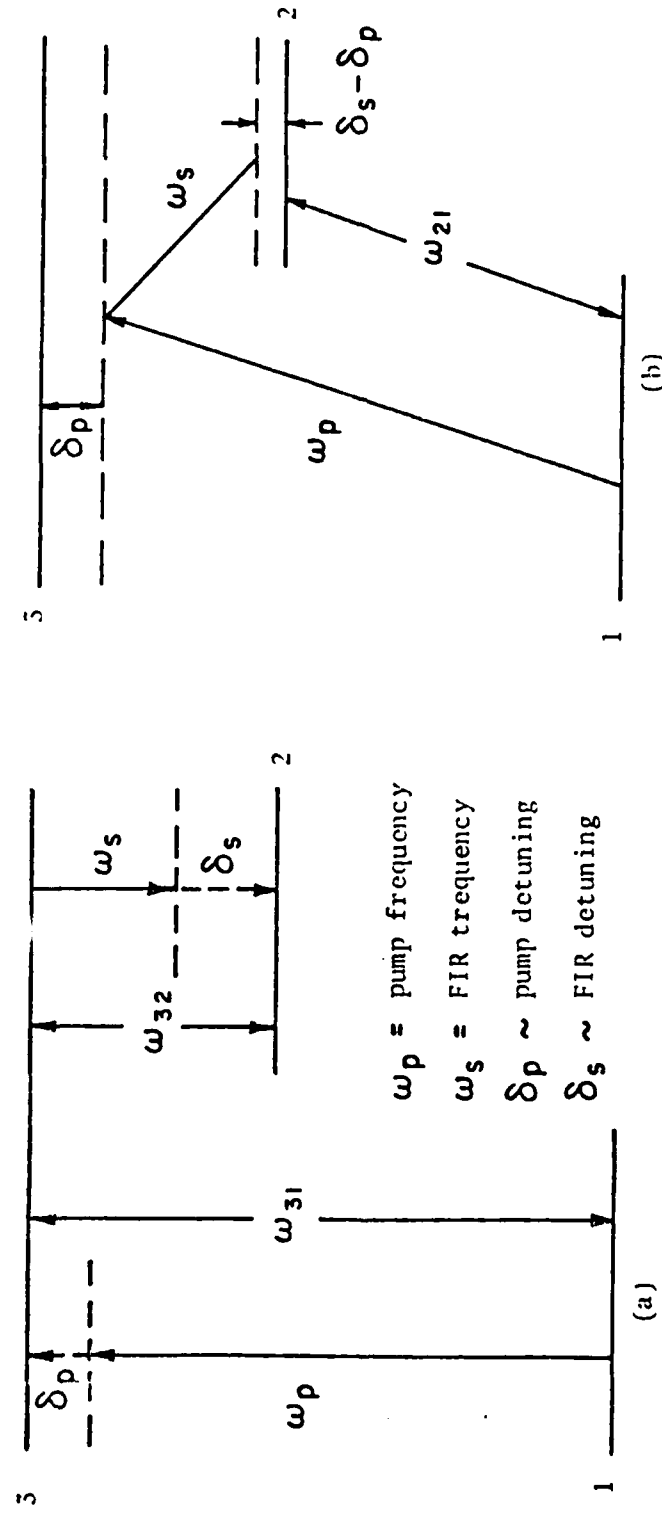


Fig. 2. Detuning Parameters (a) single photon transitions, (b) two photon transitions

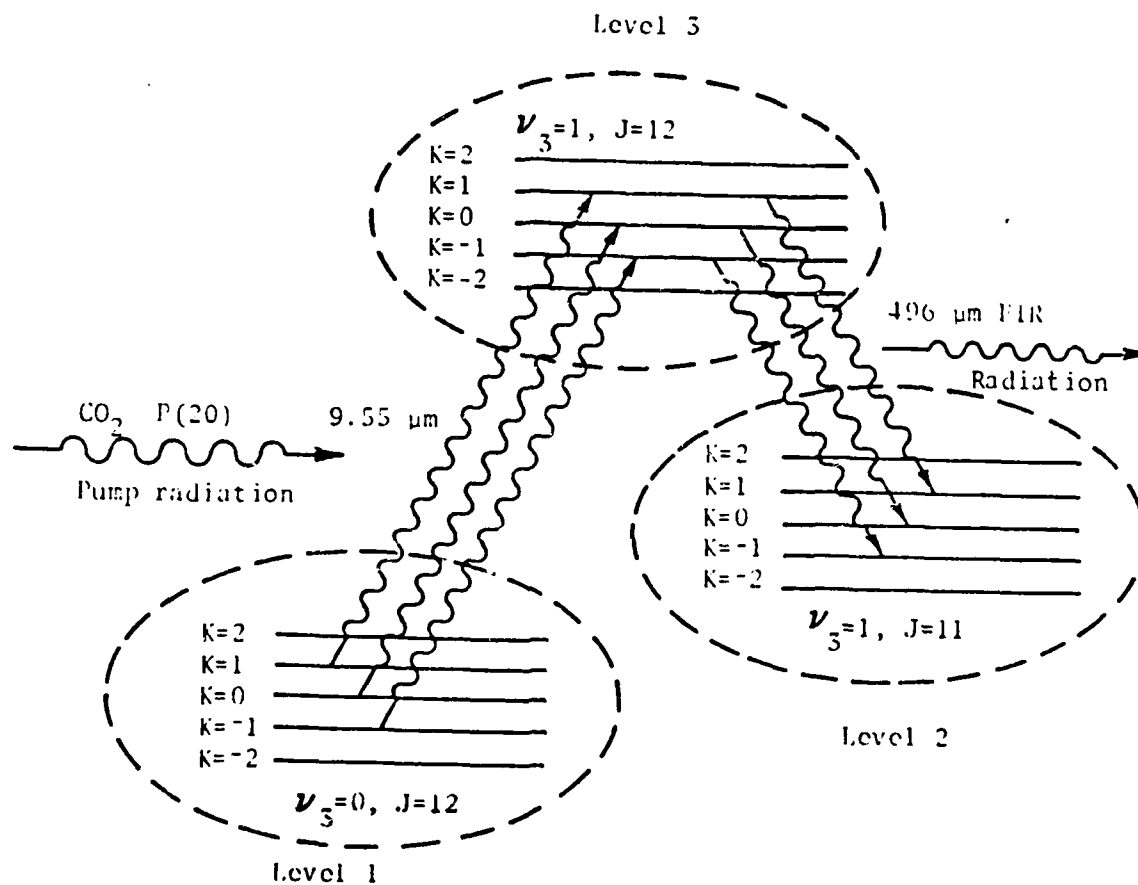


Fig. 3. Schematic of three level system.

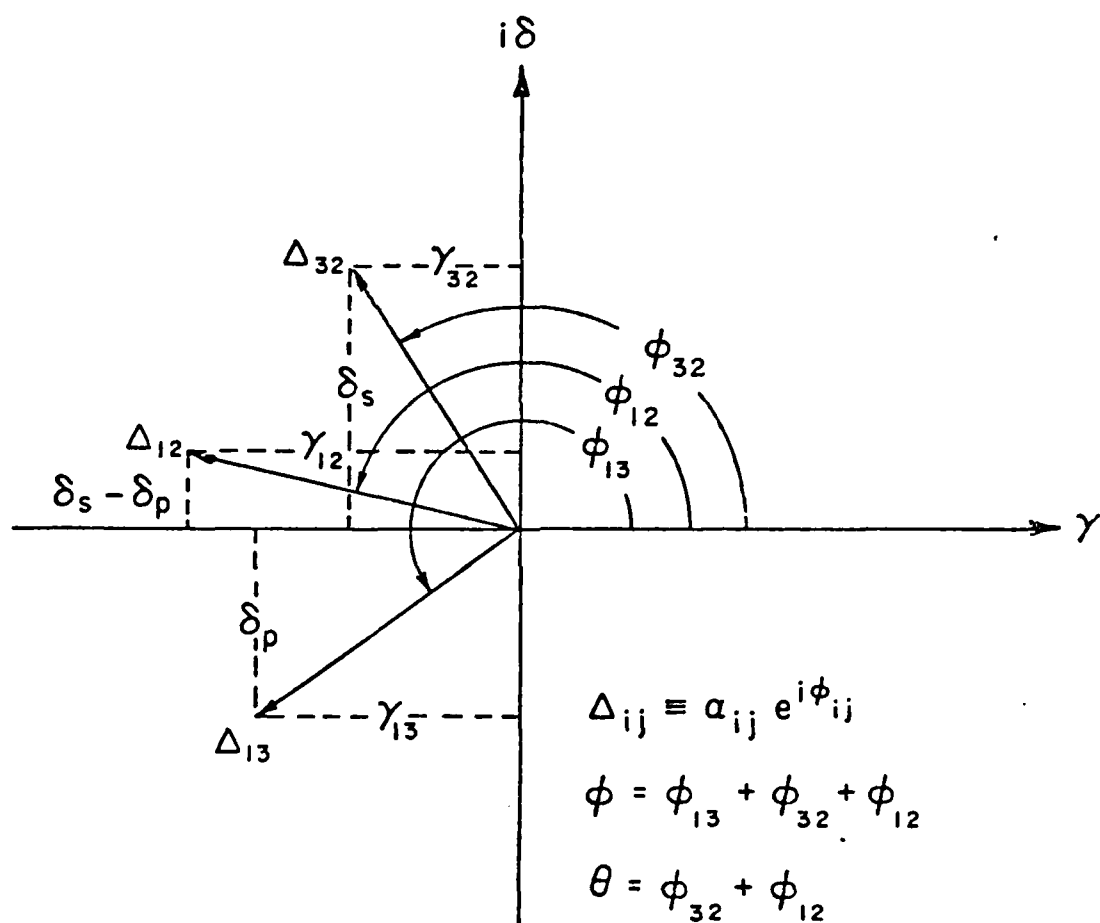


Fig. 4. Phasor Diagram of Complex Detunings.

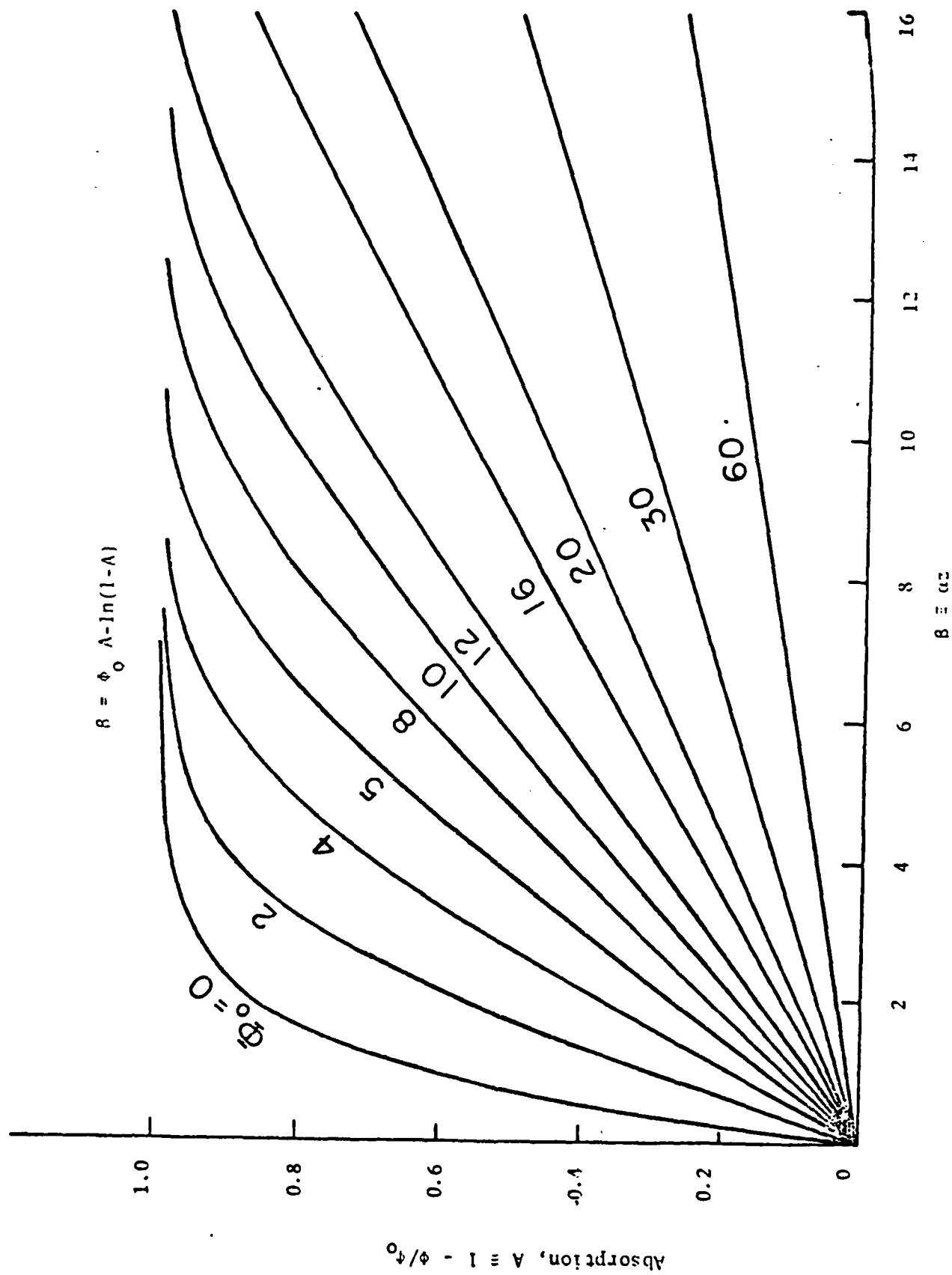


Fig. 5. Percent Absorption as function of the dimensionless absorption length αz , and the initial pump pulse area ϕ_0 .

η , Reduced FIR Gain Coefficient

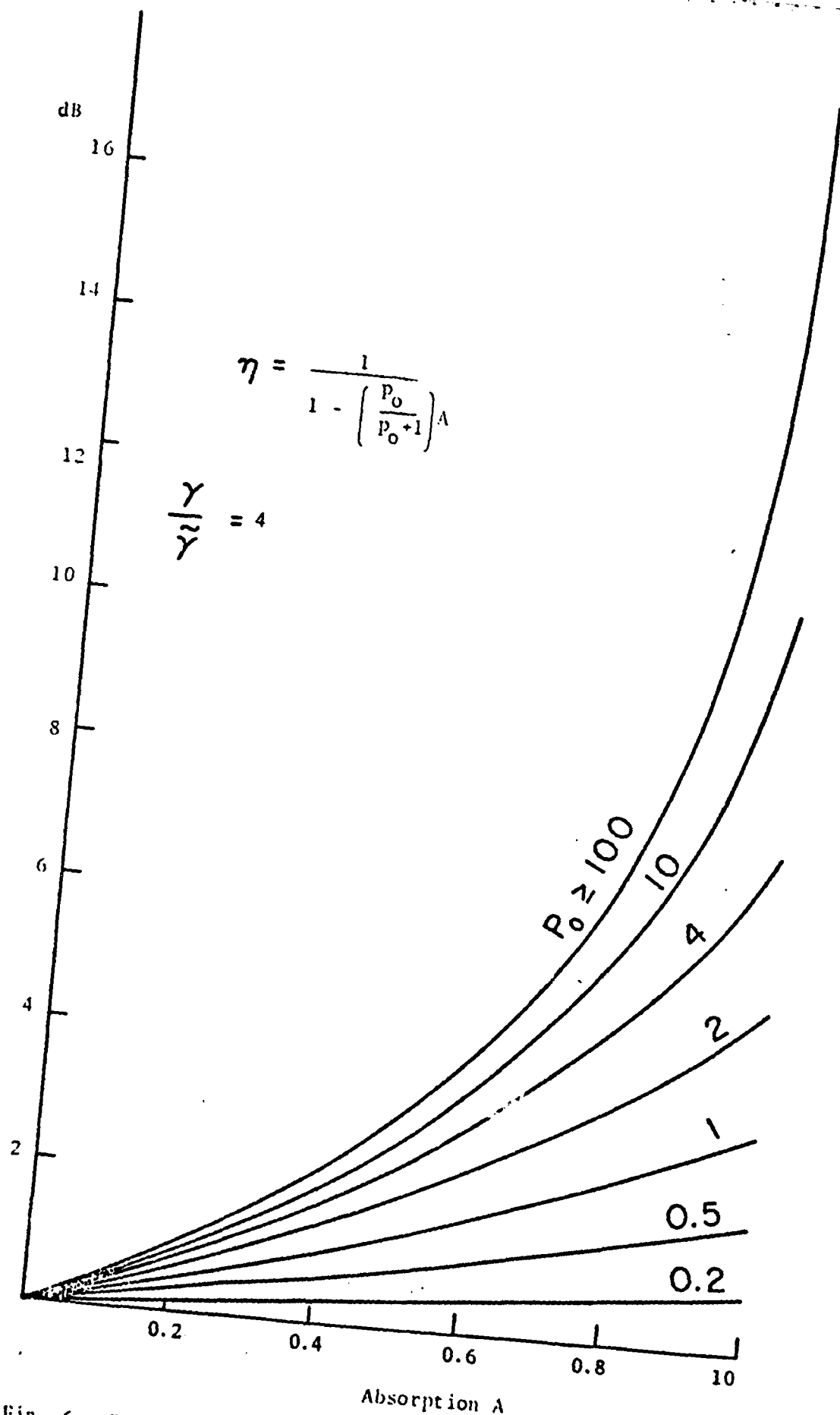


Fig. 6. Reduced FIR gain as function of percent Absorption and dimensionless pump intensity.

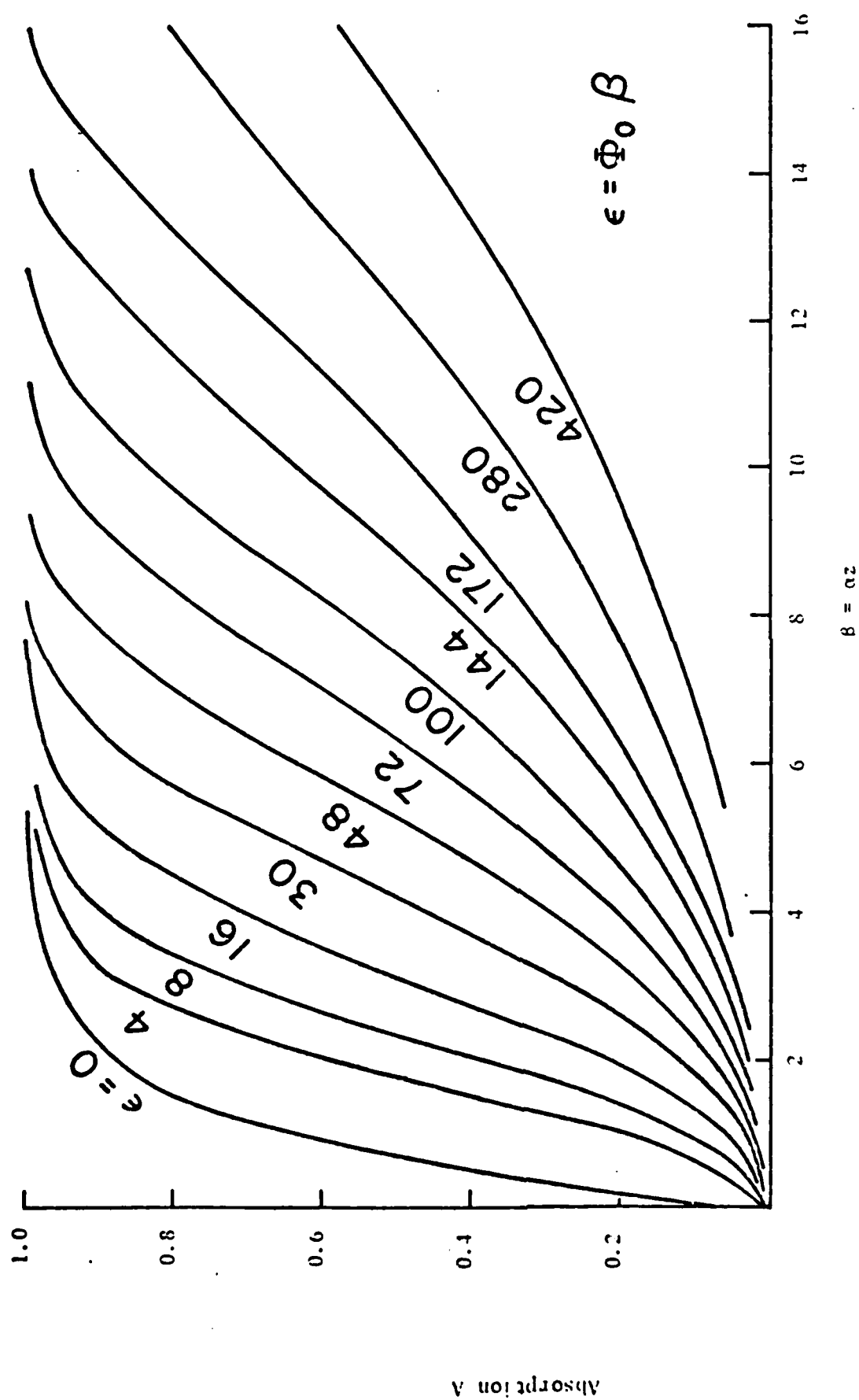


Fig. 7. Percent Absorption as function of the dimensionless absorption length αz and dimensionless power parameter ϵ .

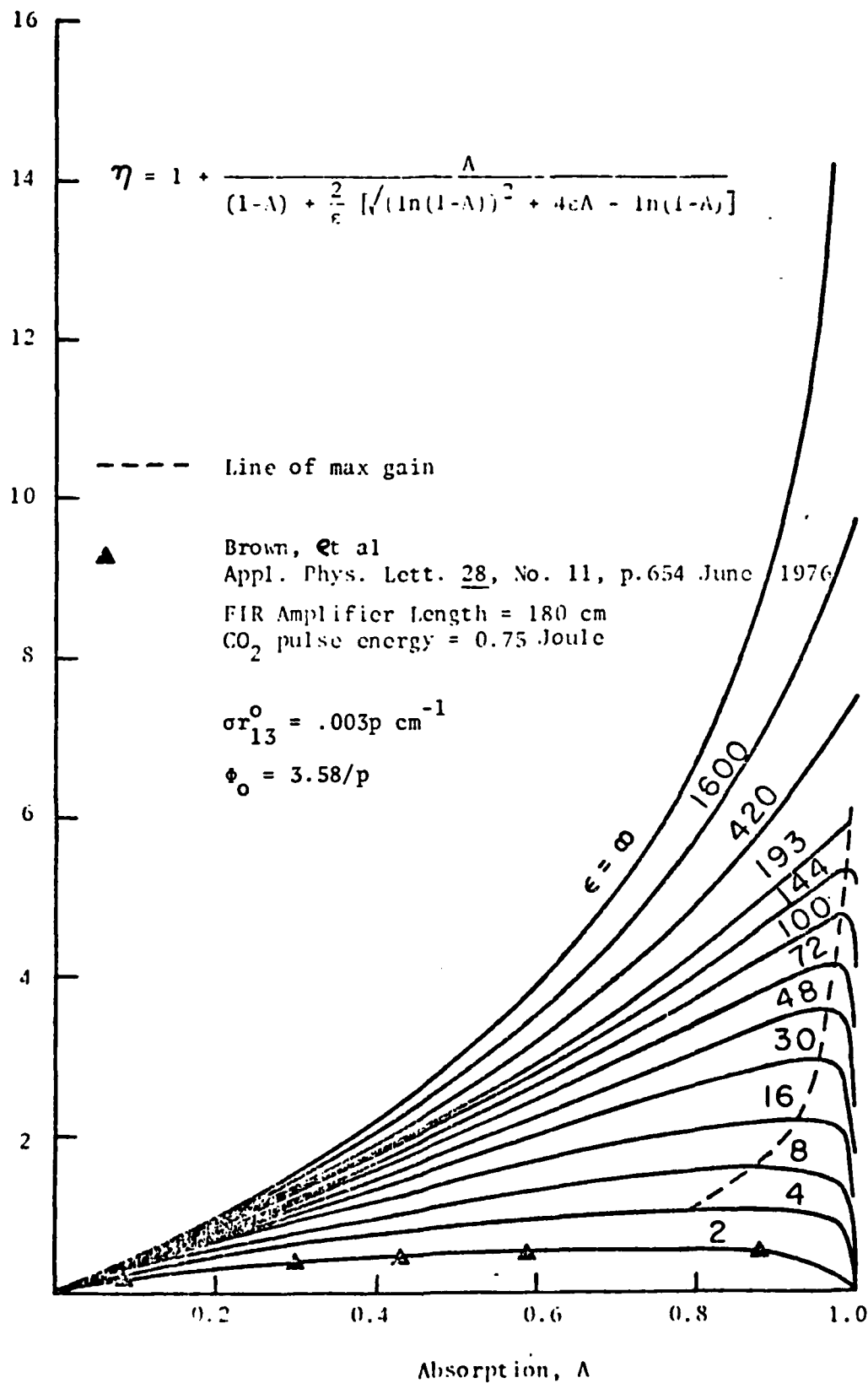


Fig. 8. Reduced FIR Gain as function of Percent Absorption and dimensionless power parameter ϵ .

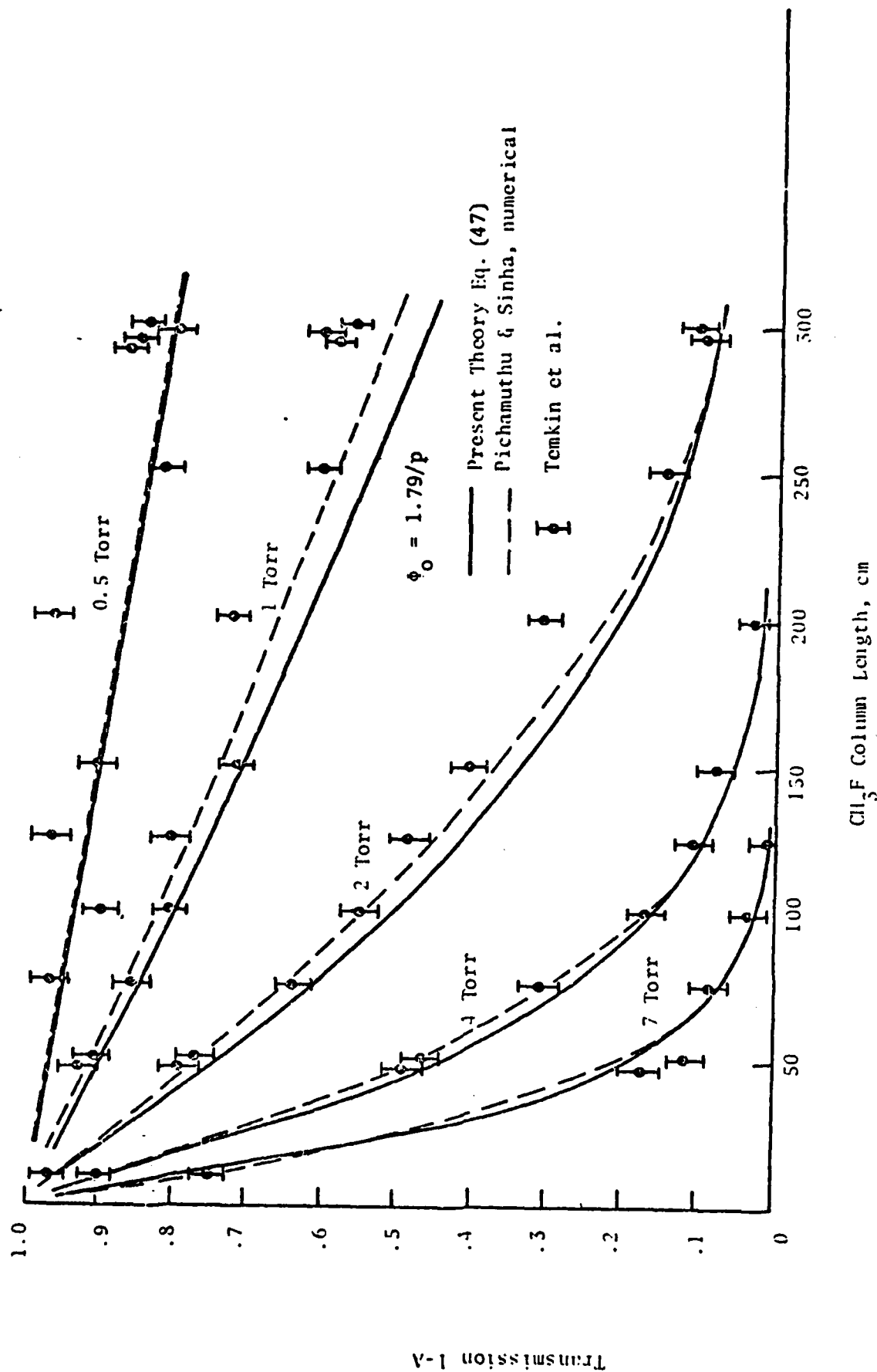


Fig. 9. Comparison between experiment,² numerical solutions⁶ and present theory, Eq. 47.

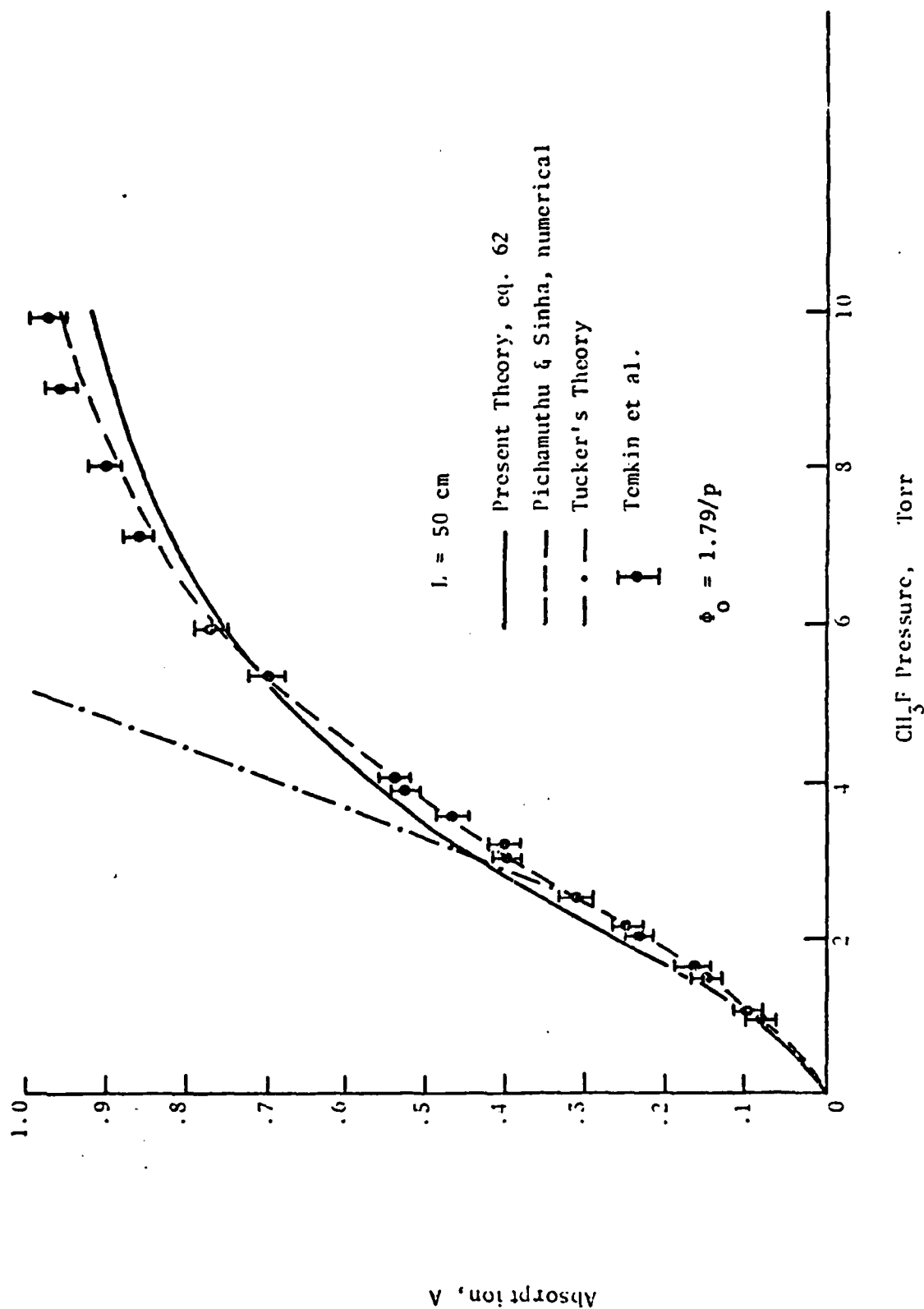


Fig. 10. Saturation Absorption. Comparison between experiment²¹ numerical solution,⁶ Tucker's theory⁴ and present theory, Eq. 47.

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