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# Strong-Field Saturation Effects in Laser Media

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Using Lamb's model, we analyze the effects of gain saturation by strong traveling-wave fields in dilute laser media. Using approximate solutions of an integral equation for the population inversion density (PID), the index of refraction and incremental gain are studied for arbitrarily strong fields. Effects of atomic motion are included for a Maxwellian velocity distribution, but pressure effects are neglected. The case of a monochromatic field leads to the saturation results of Gordon, White, and Rigden, which are studied as a function of frequency. For a small ratio of natural to Doppler linewidths, there is a transition from inhomogeneous to homogeneous broadening for sufficiently strong fields. An effect of particular interest is the generation of waves at $2\omega_1-\omega_2$, $2\omega_2-\omega_1$, and higher order sidebands by two strong input signals at $\omega_1$ and $\omega_2$. The source of the parametric gain at these frequencies is the time-dependent gain saturation due to the presence of multiple strong fields. For $\omega_1-\omega_2$ small compared to the decay rates $\gamma_a$ and $\gamma_b$ of the laser levels, the gain at these intermodulation sidebands is computed as a function of the field strengths. The limiting cases of homogeneous and inhomogeneous (due to atomic motion) broadening are studied in detail. These two cases give essentially the same results, for a given unsaturated gain. Numerical results indicate that a first-order side-band intensity at least 10% as large as that of the inducing fields can be easily observed in practice. The integral equation for the PID is converted into an infinite set of linear algebraic equations for a typical solid-state laser. The conditions under which this set of equations can be limited to a finite number is discussed, and the dependence of the sideband gain on $\omega_1-\omega_2$ is calculated. For large $\omega_1-\omega_2$, the gain at the $m$th sideband decreases as $(\omega_1-\omega_2)^{-m}$.

## I. INTRODUCTION

THE characteristics of laser oscillators have been studied in detail by Lamb.[1,2] He concentrated on the case of a strongly Doppler-broadened gaseous medium. Other authors[3–9] have given similar results with extensions to allow treatment of pressure effects,[5–7] the traveling-wave laser oscillator,[8] and the application of a magnetic field to the medium.[9]

In this work, we study the behavior of traveling waves in dilute laser amplifying media, using a formalism essentially the same as that of Lamb.[1,2] The index of refraction, incremental gain, and population inversion density (PID) are studied as functions of frequency and field strength for steady-state conditions with constant and uniform excitation of the medium. We are particularly interested in treating the situation where the fields are strong enough to render a perturbation solution useless, especially for the case of interacting waves. The use of traveling waves helps in identifying the physical processes occurring as parametric effects with damping. The powerful concept of "hole burning," introduced by Bennett[10,11] for gaseous lasers, is used in discussin g and interpreting the results.

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## II. THE EQUATIONS OF MOTION

In this section, we derive the equations describing the electromagnetic field and the active atoms of the medium. The model and approach are essentially those of Lamb's two-level theory,[1,2] and will therefore not be discussed in detail.

A partial expansion of the wave function in the two orthonormal laser states $|a)$ and $|b)$ with energies $\hbar\omega_a>\hbar\omega_b$ is

$$\psi=a(t)|a)+b(t)|b). \tag{1}$$

Substitution of (1) into the time-dependent Schrödinger equation and neglect of the small perturbation term proportional to $A^2$ leads to the equations

$$\dot{\rho}_{ab}=-(\gamma+i\omega_0)\rho_{ab}+(\rho_{aa}-\rho_{bb})(\omega_0\mathbf{A}\cdot\mathbf{P}_0/\hbar), \tag{2}$$

$$\dot{\rho}_{aa}=-\gamma_a\rho_{aa}-(\rho_{ab}+\rho_{ab}^*)(\omega_0\mathbf{A}\cdot\mathbf{P}_0/\hbar), \tag{3}$$

and

$$\dot{\rho}_{bb}=-\gamma_b\rho_{bb}+(\rho_{ab}+\rho_{ab}^*)(\omega_0\mathbf{A}\cdot\mathbf{P}_0/\hbar), \tag{4}$$

where $\rho_{ab}=ab^*$, $\rho_{aa}=aa^*$, $\rho_{bb}=bb^*$, $\omega_0=\omega_a-\omega_b$, $\gamma=(\gamma_a+\gamma_b)/2$ and $\gamma_a$ and $\gamma_b$ are the phenomenologically introduced decay rates of the upper and lower levels. $\mathbf{A}$ is the vector potential of the electromagnetic field[12] and

$$\mathbf{P}_0=-e(a|\mathbf{r}|b). \tag{5}$$

[12] The vector potential is used here in the Coulomb gauge [see, for example, L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), Chap. X]. A better approach would be to make the electric dipole approximation and transform from the Coulomb gauge to a gauge in which the electric field appears naturally in the perturbation. Using the latter gauge, in which the $\omega_0/\omega$ factors do not appear, is physically more meaningful [see, for example, E. A. Power and S. Zienau, Phil. Trans. Roy. Soc. **A251**, 427 (1959) and their references]. Since the factors $\omega_0/\omega$ are nearly unity, the present use of the vector potential is equivalent for the purposes of this paper to Lamb's use of the electric field (Refs. 1 and 2).

The classical treatment of the electromagnetic field requires $\rho_{ab}$ for the calculation of the macroscopic polarization of the medium, which is taken to be the sum over all atoms per unit volume of the microscopic dipole moment

$$\mathbf{P}=(\psi|-e\mathbf{r}|\psi)=\mathbf{P}_0(\rho_{ab}+\rho_{ab}{}^*). \qquad (6)$$

Since we will be interested here only in fields linearly polarized along the same direction, we will take $\mathbf{P}_0$ to be parallel to $\mathbf{E}$. For more general fields, one should explicitly take into account the several magnetic sublevels of one or both of the laser levels.

We write the electric field as a sum of traveling waves,

$$\mathbf{E}(r,t)=\sum_\omega \cos(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi_\omega), \qquad (7)$$

where $\mathbf{E}_\omega$, $\mathbf{k}$, and $\phi_\omega$ are the spatially dependent amplitude, propagation vector and phase of the wave with frequency $\omega$. To be completely general, the summation in Eq. (7) should be an integral over frequency components. However, we wish to approach the problem of an amplifier with a signal consisting of a small number of discrete frequency components incident on it. In writing Eq. (7) we have assumed that the amplified signal still consists of a set of discrete frequency components, although the number of components may be much larger. This approach conforms closely with experiments, and the results are in good agreement with experiments (see Ref. 13).

From (7) and $\mathbf{E}=-\partial\mathbf{A}/\partial t$, we can write the vector potential as

$$\mathbf{A}(\mathbf{r},t)=\sum_\omega (\mathbf{E}_\omega/\omega)\sin(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi_\omega). \qquad (8)$$

Thus the equations of motion (2) through (4) become

$$\dot\rho_{aa}=-\gamma_a\rho_{aa}-(\rho_{ab}+\rho_{ab}{}^*)\frac{\omega_0}{\hbar}\sum_\omega\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\sin(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi_\omega), \qquad (9)$$

$$\dot\rho_{bb}=-\gamma_b\rho_{bb}+(\rho_{ab}+\rho_{ab}{}^*)\frac{\omega_0}{\hbar}\sum_\omega\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\sin(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi_\omega), \qquad (10)$$

and

$$\dot\rho_{ab}=-(\gamma+i\omega_0)\rho_{ab}+(\rho_{aa}-\rho_{bb})\frac{\omega_0}{\hbar}\sum_\omega\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\times\sin(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi_\omega). \qquad (11)$$

In a medium with specific polarization $\mathbf{P}(\mathbf{r},t)$, Maxwell's equations give the wave equation (in mks units)

$$-\nabla^2\mathbf{E}(\mathbf{r},t)+\frac{1}{c^2}\frac{\partial^2\mathbf{E}(\mathbf{r},t)}{\partial t^2}=-\frac{1}{\epsilon_0c^2}\frac{\partial^2\mathbf{P}(\mathbf{r},t)}{\partial t^2}. \qquad (12)$$

The $\mathbf{E}_\omega$, $\mathbf{k}$, and $\phi_\omega$ in (7) are functions of $\mathbf{r}$ in the medium. Second-order spatial derivatives of these quantities can be neglected if their variation is small enough, requiring

$$\alpha\ll\omega/c\approx10^6\ \mathrm{m}^{-1} \qquad (13)$$

for optical frequencies, where $\alpha$ is the linear exponential field gain due to the medium.

Using the form

$$\mathbf{P}(\mathbf{r},t)=\sum_\omega \mathbf{P}_{\omega c}\cos(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi_\omega)+\mathbf{P}_{\omega s}\times\sin(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi_\omega) \qquad (14)$$

for the polarization, (12) reduces to the equations

$$[n(\omega)-1+(c/\omega)\mathbf{e}_\omega\cdot\nabla\phi_\omega]E_{\omega j}=(1/2\epsilon_0)P_{\omega cj}, \qquad (15)$$

and

$$\mathbf{e}_\omega\cdot\nabla E_{\omega j}=(\omega/2\epsilon_0c)P_{\omega sj}, \qquad (16)$$

where

$$\mathbf{k}(\omega)=n(\omega)\omega\mathbf{e}_\omega/c, \qquad (17)$$

and $j$ denotes the field components transverse to $\mathbf{k}$. According to Eq. (15) the in-phase component of the polarization can be viewed as contributing either to $n(\omega)$ or $\nabla\phi_\omega$. In this work we have arbitrarily assigned to $n(\omega)$ the part which is independent of distance along the direction of propagation (except for an implicit dependence through the field amplitudes), and retained $\phi_\omega$ to describe a fluctuating phase. Since only the former is present when the propagation mismatch between induced field and inducing polarization is neglected, the fluctuating phase will not concern us here.[14]

Occasionally (for example, in the oscillator problems treated by Lamb[1,2]), one wishes to consider fields whose amplitudes vary with time rather than in space. The equations for such fields are derived in an identical way, and are given by

$$\left[n(\omega)-1+\frac{1}{\omega}\frac{d\phi_\omega}{dt}\right]E_{\omega j}=\frac{1}{2\epsilon_0}P_{\omega cj}, \qquad (18)$$

and

$$(dE_{\omega j}/dt)=(\omega/2\epsilon_0)P_{\omega sj}. \qquad (19)$$

In order to take into account gaseous media, we note that an atom moving uniformly with a velocity $\mathbf{v}$, initially at $\mathbf{r}_0$ at time $t_0$, has the position $\mathbf{r}=\mathbf{r}_0+\mathbf{v}(t-t_0)$ at time $t$. We neglect pressure effects, which in many cases are small.

The quantities $\rho_{aa}$, $\rho_{bb}$, and $\rho_{ab}$ are functions of time, and for given fields the solutions are defined by the initial conditions. The simplest initial condition placed on a single atom is that it is either in state $|a)$ or $|b)$ at $t_0$, and no other possibilities will be considered here. We define the excitation process in terms of the number of atoms excited to the states $|a)$ and $|b)$ per unit time, per unit volume, per unit velocity interval. We assume that this number is a function only of velocity, and that the excitation velocity distribution is the same for both levels. The excitation is thus defined by $\lambda_{a,b}W(\mathbf{v})$, where

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[14] D. H. Close, Scientific Report No. 5 under Air Force Contract AF49(638)-1322, 1965 (unpublished); Ph.D. thesis, California Institute of Technology, 1965 (unpublished).

$\lambda_{a,b}$ are the total number of atoms excited to $|a\rangle$, $|b\rangle$ per second per meter$^3$, and $W(\mathbf{v})$ is the normalized velocity distribution.

The required solution at time $t$ is found by summing over all previous excitation times the solutions due to individual excitations. For example, if we denote the solution of (3) for atoms excited to state $|a\rangle$ at time $t_0$ and position $\mathbf{r}_0$ with velocity $\mathbf{v}$ as

$$\rho_{aa}^{(a)}(\mathbf{r},t,\mathbf{v},\mathbf{r}_0,t_0)\,, \tag{20}$$

then we can only have contributions to the specific quantity $\rho_{aa}(\mathbf{r},t,\mathbf{v})$ for $\mathbf{r}_0$ and $\mathbf{v}$ such that $\mathbf{r}=\mathbf{r}_0+\mathbf{v}(t-t_0)$. Thus the contribution of solutions (20) to $\rho_{aa}(\mathbf{r},t,\mathbf{v})$ is given by the integral

$$\rho_{aa}^{(a)}(\mathbf{r},t,\mathbf{v})=\lambda_a W(\mathbf{v}) \times\int_{-\infty}^{t} dt_0\rho_{aa}^{(a)}(\mathbf{r}=\mathbf{r}_0+\mathbf{v}(t-t_0),\ t,\ \mathbf{v},\ \mathbf{r}_0,\ t_0)\,. \tag{21}$$

There are similar contributions to $\rho_{bb}(\mathbf{r},t,\mathbf{v})$ and $\rho_{ab}(\mathbf{r},t,\mathbf{v})$, and other contributions due to excitation to the lower level. For the field equations we of course need only $\rho_{ab}(\mathbf{r},t,\mathbf{v})$, but it is convenient and physically interesting to express this in terms of

$$\rho_{aa}(\mathbf{r},t,\mathbf{v})-\rho_{bb}(\mathbf{r},t,\mathbf{v})\equiv N(\mathbf{r},t,\mathbf{v})\,, \tag{22}$$

which is the population inversion density at $\mathbf{r},t$ of atoms with velocity $\mathbf{v}$.

In order to derive an integral equation for $N(\mathbf{r},t,\mathbf{v})$, let us denote

$$\rho_{aa}^{(a)}(\mathbf{r}=\mathbf{r}_0+\mathbf{v}(t-t_0),\ t,\ \mathbf{v},\ \mathbf{r}_0,\ t_0)$$

by $\rho_{aa}(t,t_0)$ with similar definitions of $\rho_{bb}(t,t_0)$ and $\rho_{ab}(t,t_0)$. These quantities satisfy the differential equations (9)–(11), with the initial conditions

$$\rho_{aa}(t_0,t_0)=1,\quad \rho_{bb}(t_0,t_0)=\rho_{ab}(r_0,t_0)=0\,. \tag{23}$$

Formal integration of these equations gives

$$\rho_{aa}(t,t_0)=\exp(-\gamma_a(t-t_0))-\int_{t_0}^{t}dt'\ \exp(\gamma_a(t'-t))(\rho_{ab}(t',t_0)+\rho_{ab}^*(t',t_0))\frac{\omega_0}{\hbar}\sum_{\omega}\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\right) \times\sin(\mathbf{k}\cdot\mathbf{r}_0+\mathbf{k}\cdot\mathbf{v}(t'-t_0)-\omega t'+\phi)\,, \tag{24}$$

$$\rho_{bb}(t,t_0)=\int_{t_0}^{t}dt'\ \exp(\gamma_b(t'-t))(\rho_{ab}(t',t_0)+\rho_{ab}^*(t',t_0))\frac{\omega_0}{\hbar}\sum_{\omega}\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\right)\sin(\mathbf{k}\cdot\mathbf{r}_0+\mathbf{k}\cdot\mathbf{v}(t'-t_0)-\omega t'+\phi)\,, \tag{25}$$

and

$$\rho_{ab}(t,t_0)=\int_{t_0}^{t}dt'\ \exp[(\gamma+i\omega_0)(t'-t)](\rho_{aa}(t',t_0)-\rho_{bb}(t',t_0))\frac{\omega_0}{\hbar}\sum_{\omega}\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\right)\sin(\mathbf{k}\cdot\mathbf{r}_0+\mathbf{k}\cdot\mathbf{v}(t'-t_0)-\omega t'+\phi)\,. \tag{26}$$

We can subtract (25) from (24) to obtain an expression for $\rho_{aa}(t,t_0)-\rho_{bb}(t,t_0)$ in terms of $\rho_{ab}(t',t_0)$. Substituting $\rho_{ab}(t',t_0)$ from (26) into this expression gives an integral equation for $\rho_{aa}(t,t_0)-\rho_{bb}(t,t_0)$. If we sum over all atoms per unit volume at $\mathbf{r}_0$, we obtain a factor of $\lambda_a W(\mathbf{v})$. Integrating over all possible excitation times $t_0$ and using (21), we find

$$\rho_{aa}^{(a)}(t,t_0)-\rho_{bb}^{(a)}(t,t_0)=\lambda_a W(\mathbf{v})/\gamma_a-\lambda_a W(\mathbf{v})(\omega_0/2\hbar)^2\int_{-\infty}^{t}dt_0\int_{t_0}^{t}dt'\int_{t_0}^{t'}dt''(e^{\gamma_a(t'-t)}+e^{\gamma_b(t'-t)}) \times(e^{(\gamma+i\omega_0)(t''-t')}+e^{(\gamma-i\omega_0)(t''-t')})(\rho_{aa}^{(a)}(t'',t_0)-\rho_{bb}^{(a)}(t'',t_0))\sum_{\omega,\omega'}\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\right)\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_{\omega'}}{\omega'}\right) \times[\exp i[(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}-\mathbf{k}\cdot\mathbf{v}(t-t')+\mathbf{k}'\cdot\mathbf{v}(t-t'')-\omega t'+\omega' t''+\phi-\phi']+\text{c.c.}]\,, \tag{27}$$

where we have neglected terms of relative order $\gamma/\omega_0$ and used $\mathbf{r}=\mathbf{r}_0+\mathbf{v}(t-t_0)$.

In the same way, we find a similar integral equation for the contribution to $N(\mathbf{r},t,\mathbf{v})$ due to excitation of the state $|b\rangle$. Adding these two contributions, interchanging the integration order twice, and performing the $t_0$ integration, we obtain

$$N(\mathbf{r},t,\mathbf{v})=N_0W(\mathbf{v})-\left(\frac{\omega_0}{2\hbar}\right)^2\int_0^\infty dt_1\int_0^\infty dt_2(\exp(-\gamma_a t_1)+\exp(-\gamma_b t_1))N(\mathbf{r}\cdot\ t-t_1-t_2,\ \mathbf{v})\sum_{\omega,\omega'}\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\right)\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_{\omega'}}{\omega'}\right) \times[\exp[i\Delta+i(\omega-\omega'-(\mathbf{k}-\mathbf{k}')\cdot\mathbf{v})t_1-(\gamma-i(\omega_0-\omega'+\mathbf{k}'\cdot\mathbf{v}))t_2]+\text{c.c.}]\,, \tag{28}$$

where $t_1=t-t'$, $t_2=t'-t''$, $N_0=\lambda_a/\gamma_a-\lambda_b/\gamma_b$, and

$$\Delta=(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}-(\omega-\omega')t+\phi-\phi'\,. \tag{29}$$

Equation (28) is the integral equation for the population inversion density. We can express $\rho_{ab}(\mathbf{r},t,\mathbf{v})$ in terms of $N(\mathbf{r},t,\mathbf{v})$ by starting from (26). In an identical way we find

$$\rho_{ab}(\mathbf{r},t,\mathbf{v})=\frac{\omega_0}{2i\hbar}\int_0^\infty dt_1 N(\mathbf{r},\,t-t_1,\,\mathbf{v})\sum_\omega\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\right)\exp[i(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi)-(\gamma+i(\omega_0-\omega+\mathbf{k}\cdot\mathbf{v}))t_1]. \tag{30}$$

## III. SOLUTIONS

The integral equation (28) usually cannot be solved exactly. The zero-field solution is obviously

$$N(\mathbf{r},t,\mathbf{v})=N_0W(\mathbf{v}), \tag{31}$$

so that $N(\mathbf{r},t)=N_0$. If the fields are weak enough, (28) can be iterated to form a series solution in powers of the fields. The linear solution is obtained by substituting (31) into (30). The lowest order nonlinear solution is obtained by inserting (31) into the right-hand side of (28); the resulting second order $N(\mathbf{r},t,\mathbf{v})$ gives a third order $\rho_{ab}$ (hence polarization) upon substitution into (30). Higher order terms of the solution can be obtained by further iterations.

The iterative solution will be valid if the fields are always small compared with a field determined by the system parameters. This field is given by[14]

$$E_0^2=\gamma_a\gamma_b\hbar^2/P_0^2. \tag{32}$$

The corresponding intensity, $I_0=\epsilon_0 cE_0^2$ is the same as the saturation intensity occurring in treatments of laser saturation.[15–17] If two fields comparable to or larger than $E_0$ interact, the results could at best be predicted qualitatively from the series solution by consideration of higher order terms of the series. In order to study such a situation more accurately, we must find other approximate solutions for the integral equation (28).

Examining (28), we see that fields at two different frequencies will induce a variation in $N(\mathbf{r},t,\mathbf{v})$ at their difference frequency. Also, we see that the solution is characterized by the rate of variation of $N(\mathbf{r},t,\mathbf{v})$ compared to $\gamma_a$ and $\gamma_b$–rates slow compared to $\gamma_a$, $\gamma_b$ will appear strongly, while rates very rapid compared to $\gamma_a$, $\gamma_b$ will have small amplitude. However, if $N(\mathbf{r},t,\mathbf{v})$ varies slowly compared to $\gamma_a$, $\gamma_b$, the integrations will be dominated by the $[\exp(-\gamma_a t_1)+\exp(-\gamma_b t_1)]$ and $\exp(-\gamma t_2)$ factors, and $N(\mathbf{r},t,\mathbf{v})$ may be removed from the integral, giving upon integration,

$$N(\mathbf{r},t,\mathbf{v})=N_0W(\mathbf{v})\Bigg\{1+\sum_{\omega,\omega'}\left(\frac{\omega_0}{2\hbar}\right)^2\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\right)\left(\frac{\mathbf{P}_0\cdot\mathbf{E}_{\omega'}}{\omega'}\right)[\exp(i\Delta)/(\gamma-i(\omega_0-\omega'+\mathbf{k}'\cdot\mathbf{v})) \times[1/(\gamma_a-i(\omega-\omega'-(\mathbf{k}-\mathbf{k}')\cdot\mathbf{v}))+1/[\gamma_b-i(\omega-\omega'-(\mathbf{k}-\mathbf{k}')\cdot\mathbf{v})]]+\text{c.c.}]\Bigg\}^{-1}. \tag{33}$$

According to the above discussion, we should neglect terms in the summation for which $|\omega-\omega'|\gg\gamma_a,\gamma_b$ and keep the terms for which $|\omega-\omega'|\ll\gamma_a,\gamma_b$. For intermediate frequency separations, (33) will be only qualitatively correct. This result shows that the PID will contain all harmonics of each frequency difference $\omega-\omega'$ which are small compared to $\gamma_a$, $\gamma_b$, and will contain higher harmonics to a lesser degree than is indicated by (33).

Since $N(\mathbf{r},t,\mathbf{v})$ is slowly varying compared to $\gamma$, we may take it outside the integral in (30), giving

$$\rho_{ab}(\mathbf{r},t,\mathbf{v})=\frac{\omega_0}{2i\hbar}N(\mathbf{r},t,\mathbf{v})\sum_\omega\frac{\mathbf{P}_0\cdot\mathbf{E}_\omega}{\omega}\exp(i(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi))(\gamma+i(\omega_0-\omega+\mathbf{k}\cdot\mathbf{v}))^{-1}. \tag{34}$$

Using (33) in (34) gives an approximate solution, subject to the above restrictions. For our purposes we will limit it to the simplest and most interesting case of single-polarization waves traveling in the same direction, so that $\mathbf{k}-\mathbf{k}'\approx 0$. We therefore replace $\mathbf{P}_0(\mathbf{P}_0\cdot\mathbf{E}_\omega)$ with $E_\omega P_0^2/3$ in order to get the correct linear result, and $(\mathbf{P}_0\cdot\mathbf{E}_\omega)(\mathbf{P}_0\cdot\mathbf{E}_{\omega'})$ with $P_0^2E_\omega E_{\omega'}$, for simplicity. This will introduce small errors in the numerical factors which are within the present approximation, but will allow easier computation and emphasis on the primary effects of interest.

We thus find for the polarization

$$P(\mathbf{r},t,\mathbf{v})=\langle\mathbf{P}_0(\rho_{ab}(\mathbf{r},t,\mathbf{v})+\rho_{ab}^*(\mathbf{r},t,\mathbf{v}))\rangle\cong(\omega_0P_0^2/6i\hbar)N(\mathbf{r},t,\mathbf{v})\sum_\omega(E_\omega/\omega) \times[\exp(i(\mathbf{k}\cdot\mathbf{r}-\omega t+\phi))/(\gamma+i(\omega_0-\omega+\mathbf{k}\cdot\mathbf{v}))-\text{c.c.}], \tag{35}$$

where

$$N(\mathbf{r},t,\mathbf{v})=N_0W(\mathbf{v})\{1+\sum_{\omega',\omega''}(\gamma E_{\omega'}E_{\omega''}/2E_0^2)[\exp(i\Delta)/(\gamma-i(\omega_0-\omega''+\mathbf{k}''\cdot\mathbf{v}))+\text{c.c.}]\}^{-1}, \tag{36}$$

[15] E. I. Gordon, A. D. White, and J. D. Rigden, in *Proceedings of the Symposium on Optical Masers at the Polytechnic Institute of Brooklyn, 1963* (Polytechnic Press, Brooklyn, New York, 1963), p. 309.
[16] W. W. Rigrod, J. Appl. Phys. 34, 2602 (1963).
[17] A. D. White, E. I. Gordon, and J. D. Rigden, Appl. Phys. Letters 2, 91 (1963).

and $\Delta=(\mathbf{k}'-\mathbf{k}'')\cdot\mathbf{r}-(\omega'-\omega'')t+\phi'-\phi''$. Equations (35) and (36) form the approximate solution.

We will consider only a Maxwellian velocity distribution of excited atoms, i.e.,

$$W(\mathbf{v})=(1/\pi u^2)^{3/2}\exp(-v^2/u^2), \quad (37)$$

where $u^2=2kT/M$. The special case of stationary atoms can be obtained by letting $u$ become zero, i.e., by setting $\mathbf{v}=0$.

## IV. SINGLE-FREQUENCY FIELD

For a single-frequency field we use (37) and obtain, after integrating over the velocity components perpendicular to $\mathbf{k}$, the sine and cosine components of the polarization (14)

$$\frac{\omega}{2\epsilon_0 c}P_s=\frac{\alpha_0 E\gamma^2}{\pi^{1/2}u}\int_{-\infty}^{\infty}\frac{\exp(-V^2/u^2)dV}{\gamma^2(1+E^2/E_0^2)+(\omega_0-\omega+kV)^2}, \quad (38)$$

$$\frac{1}{2\epsilon_0}P_c=-\frac{c}{\omega}\frac{\alpha_0 E\gamma}{\pi^{1/2}u}\times\int_{-\infty}^{\infty}\frac{(\omega_0-\omega+kV)\exp(-V^2/u^2)dV}{\gamma^2(1+E^2/E_0^2)+(\omega_0-\omega+kV)^2}, \quad (39)$$

where $\alpha_0=P_0^2N_0\omega_0/(6\gamma\epsilon_0 c\hbar)$. With the definitions

$$b^2=\gamma^2(1+E^2/E_0^2)/(k_0u)^2,$$
$$x=(\omega_0-\omega)/(k_0u), \quad (40)$$
$$a=\gamma/(k_0u),$$

and

$$\alpha=a\alpha_0\pi^{1/2},$$

where $k_0=\omega_0/c$, we can write the gain and index from (15) and (16) in terms of the error function for complex arguments[18] (Ref. 19, 14) as

$$\frac{1}{E}\frac{dE}{dz}=\frac{\alpha\,\mathrm{Re}w(x+ib)}{(1+E^2/E_0^2)^{1/2}}, \quad (41)$$

$$n(\omega)=1-(c\alpha/\omega)\mathrm{Im}w(x+ib), \quad (42)$$

where Re and Im indicate the real and imaginary parts, respectively.

The parameter $a$, being the ratio of the natural to the Doppler linewidths, determines the degree of Doppler broadening of the line. If the Doppler broadening predominates sufficiently, we can expand (41) and (42) to first order in $b\ll1$ (Ref. 19, 14), giving

$$(1/E)(dE/dz)=\alpha[\exp(-x^2)/(1+E^2/E_0^2)^{1/2}-2a/\pi^{1/2}(1-2xF(x))], \quad (43)$$

$$n(\omega)=1-(c\alpha/\omega)[2F(x)/\pi^{1/2}-2a(1+E^2/E_0^2)^{1/2}x\exp(-x^2)], \quad (44)$$

[18] Defined by

$$w(z)=\exp(-z^2)\mathrm{erfc}(-iz)=\frac{2}{\pi^{1/2}}\exp(-z^2)\int_z^{\infty}\exp(-t^2)dt.$$

This is essentially the complex conjugate of the "plasma dispersion function" used by Lamb (Ref. 2).

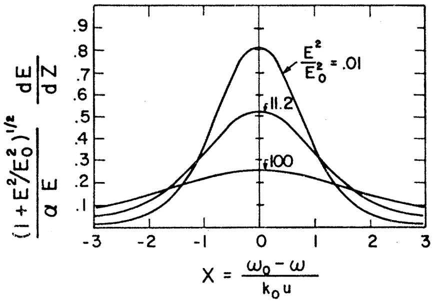


FIG. 1. Gain saturation and broadening as a function of frequency for $a=0.2$.

where

$$F(x)=\exp(-x^2)\int_0^x\exp(t^2)dt \quad (45)$$

is the Gaussian dispersion function.[19] Equations (43) and (44) are consistent with the "hole-burning" description of an inhomogeneously broadened line.[10,11] Thus the gain comes primarily from those atoms whose velocities allow them to interact strongly with the field, while the phase shift (index of refraction) arises primarily because of atoms outside this region. Correspondingly, the primary gain term is saturated, while the primary index term is not.

It should be noted that the expansion parameter above is not $a$ but $b=a(1+E^2/E_0^2)^{1/2}$, so that strong enough fields can destroy the predominance of the Doppler broadening. This is consistent with the fact that the hole or interaction linewidth increases with increasing fields. In fact, the stationary atom (homogeneous) case can be obtained either by starting from (35) and (36) with $\mathbf{v}=0$ or by using the asymptotic form of the complex error function,[19]

$$w(z)\approx i/\pi^{1/2}z \quad (46)$$

with (41) and (42), giving

$$\frac{1}{E}\frac{dE}{dz}=\frac{\alpha_0\gamma^2}{\gamma^2(1+E^2/E_0^2)+(\omega_0-\omega)^2}, \quad (47)$$

$$n(\omega)=1-\frac{c}{\omega}\frac{\alpha_0\gamma(\omega_0-\omega)}{\gamma^2(1+E^2/E_0^2)+(\omega_0-\omega)^2}. \quad (48)$$

In Fig. 1, $(1+E^2/E_0^2)^{1/2}/(\alpha E(dE/dz)$ from (41) for $a=0.2$ is plotted as a function of frequency for various saturation levels. This shows the excess saturation and broadening over the "inhomogeneous" case, given by (43). In Fig. 2 the same quantity is plotted as a function of $E^2/E_0^2$ for various values of $a$, compared to the homogeneous case, given by Eq. (47). Here a purely inhomogeneous interaction ($a=0$) would result in a horizontal

[19] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (U. S. Department of Commerce, National Bureau of Standards, Washington, D. C., 1964), Appl. Math. Ser. 55, pp. 297–304, 325–328.

line equal to 1. This indicates how any line with a finite natural linewidth becomes homogeneously broadened for sufficiently strong fields.

If the incremental gain functions (43) and (47) are expressed in terms of the intensity $I=\epsilon_0 cE^2$ for $\omega=\omega_0$, we obtain Eq. (16) and (15) of Gordon, White, and Rigden.[15] These equations can be integrated,[15–17] giving the net intensity gain of a saturated amplifier, as a function of the incident intensity. Such a set of curves is shown in Fig. 3 for the predominantly Doppler-broadened case, together with some experimental points for the 3.39-$\mu$ line in neon due to Hotz.[20]

In all cases, the above results can be expanded to lowest order in the fields, giving the linear gain and index functions and their lowest order nonlinear corrections,[14] within a small numerical error in the latter case, as discussed above.

## V. MULTIPLE-COMPONENT FIELDS

### A. Two Input Components

For the case of two waves traveling in the same direction (36) gives for $\Delta\omega\ll\gamma_a,\gamma_b$,

$$N(\mathbf{r},t,\mathbf{v})=N_0W(\mathbf{v})\left[1+\frac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{E_0^2}\times\frac{\gamma^2}{\gamma^2+(\omega_0-\omega+\mathbf{k}\cdot\mathbf{v})^2}\right]^{-1}, \quad (49)$$

where $\Delta\omega=|\omega_1-\omega_2|$ has been neglected compared to $\gamma$, $\omega=\omega_1+(\omega_2-\omega_1)/2$, $\mathbf{k}=\mathbf{k}_1+(\mathbf{k}_2-\mathbf{k}_1)/2$, and

$$\Delta_{12}=(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r}-(\omega_1-\omega_2)t+\phi_1-\phi_2. \quad (50)$$

If $\Delta\omega\gg\gamma_a,\gamma_b$, according to the discussion of (33) we can to a good approximation neglect the modulation terms, and find

$$N(\mathbf{r},t,\mathbf{v})=N_0W(\mathbf{v})\left[1+\frac{E_1^2}{E_0^2}\frac{\gamma^2}{\gamma^2+(\omega_0-\omega_1+\mathbf{k}_1\cdot\mathbf{v})^2}+\frac{E_2^2}{E_0^2}\frac{\gamma^2}{\gamma^2+(\omega_0-\omega_2+\mathbf{k}_2\cdot\mathbf{v})^2}\right]^{-1}. \quad (51)$$

In the latter case, study of the gain and index at each frequency shows the effects of saturation due to one wave on the other, for the homogeneous and inhomogeneous lines.[14] Here, we will only note that considerations of hole burning predict the saturation interaction for the inhomogeneously broadened line, and proceed to consider the former case, where the modulation terms are not neglected.

First, we compare (49) to the PID for a single wave from (36)

$$N(\mathbf{r},t,\mathbf{v})=N_0W(\mathbf{v})\left[1+\frac{E^2}{E_0^2}\frac{\gamma^2}{\gamma^2+(\omega_0-\omega+\mathbf{k}\cdot\mathbf{v})^2}\right]^{-1}. \quad (52)$$

[20] D. F. Hotz (private communication).

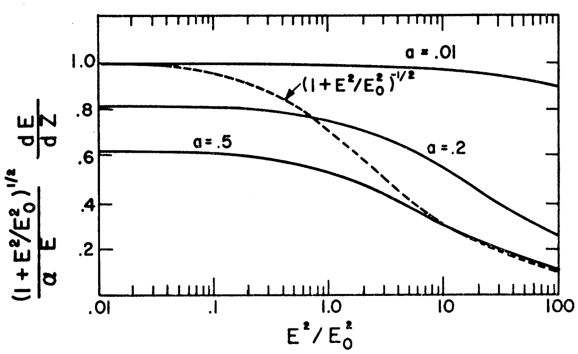


FIG. 2. Gain saturation as a function of field strength, compared to homogeneous gain saturation.

If we examine these expressions as functions of velocity, we find that (52) shows a hole whose width in frequency units is

$$2\gamma(1+E^2/E_0^2)^{1/2}, \quad (53)$$

and whose depth, relative to unity, is

$$\frac{E^2/E_0^2}{1+E^2/E_0^2}. \quad (54)$$

This hole of course appears superimposed on the distribution $W(\mathbf{v})$. Correspondingly, the two-frequency case with $\Delta\omega\gg\gamma_a,\gamma_b$ shows two similar holes in the velocity distribution of the PID, so long as $\Delta\omega$ remains large compared to the sum of the two-hole widths, i.e., the holes do not overlap. Finally, when $\Delta\omega\ll\gamma_a,\gamma_b$ (49) shows that in the limiting case where the two holes completely overlap there is again a single hole in the PID, whose width and depth vary harmonically at the frequency $\Delta\omega$. The maximum and minimum widths are

$$2\gamma[1+(E_1+E_2)^2/E_0^2]^{1/2}$$

and

$$2\gamma[1+(E_1-E_2)^2/E_0^2]^{1/2}, \quad (55)$$

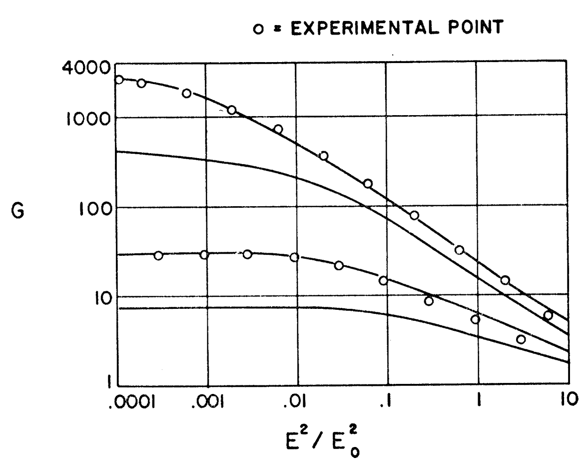


FIG. 3. Integrated gain saturation, compared with experiment.

and the corresponding depths are

$$\frac{(E_1+E_2)^2/E_0^2}{1+(E_1+E_2)^2/E_0^2} \quad\text{and}\quad \frac{(E_1-E_2)^2/E_0^2}{1+(E_1-E_2)^2/E_0^2}. \tag{56}$$

This fluctuating hole shows how the PID follows the beating of the two incident fields, for $\Delta\omega \ll \gamma_a,\gamma_b$. The PID is modulated at all harmonics of $\Delta\omega$, the modulation taking the form of waves traveling through the medium. It is the parametric interaction of the fields with these modulation waves which generates new fields at the various "intermodulation" or "combination tone" frequencies, the higher modulation harmonics giving rise to higher order combination tones, as we shall now see.

Substituting (49) into (35), we obtain the polarization for a Maxwellian velocity distribution of excited atoms,

$$\frac{\omega P(\mathbf{r},t)}{2\epsilon_0 c}=\frac{\alpha}{\pi}\int_{-\infty}^{\infty}[\exp(-y^2)dy]\Big/\left\{a^2\left[1+\frac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{E_0^2}\right]+(x+y)^2\right\}$$
$$\times[aE_1\sin(\mathbf{k}_1\cdot\mathbf{r}-\omega_1t+\phi_1)-(x_1+y)E_1\cos(\mathbf{k}_1\cdot\mathbf{r}-\omega_1t+\phi_1)$$
$$+aE_2\sin(\mathbf{k}_2\cdot\mathbf{r}-\omega_2t+\phi_2)-(x_2+y)E_2\cos(\mathbf{k}_2\cdot\mathbf{r}-\omega_2t+\phi_2)], \tag{57}$$

where the two velocity integrations perpendicular to $\mathbf{k}_i$ have been carried out, $x=(\omega_0-\omega)/(k_0u)$ and $y=V/u$. This can be expressed in terms of the complex error function, but we will study only two limiting cases, first, stationary atoms or the homogeneous line and second, the inhomogeneous line.

For stationary atoms, we find either from the limiting form of the complex error function or from starting with $\mathbf{v}=0$

$$\frac{\omega P(\mathbf{r},t)}{2\epsilon_0 c}=\alpha_0\gamma\Big/\left\{\gamma^2\left[1+\frac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{E_0^2}\right]+(\omega_0-\omega)^2\right\}$$
$$\times[\gamma E_1\sin(\mathbf{k}_1\cdot\mathbf{r}-\omega_1t+\phi_1)-(\omega_0-\omega_1)E_1\cos(\mathbf{k}_1\cdot\mathbf{r}-\omega_1t+\phi_1)$$
$$+\gamma E_2\sin(\mathbf{k}_2\cdot\mathbf{r}-\omega_2t+\phi_2)-(\omega_0-\omega_2)E_2\cos(\mathbf{k}_2\cdot\mathbf{r}-\omega_2t+\phi_2)]. \tag{58}$$

The polarization thus contains the frequencies $\omega_1\pm m\Delta\omega$ for integral $m$. The Fourier analysis in the Appendix gives the polarization at each frequency. We find

$$\omega P_s(\omega_1)/(2\epsilon_0c)=\alpha_0[E_1C_0+E_2C_1/2], \tag{59}$$

$$\omega P_c(\omega_1)/(2\epsilon_0c)=-\alpha_0(\omega_0-\omega)/\gamma[E_1C_0+E_2C_1/2], \tag{60}$$

with a similar expression for $P(\omega_2)$, and at $\omega_{\pm m}=\omega_1\pm m\times(\omega_1-\omega_2)\neq\omega_1,\omega_2$ the magnitude of the polarization due to $E_1$ and $E_2$ is

$$\omega P_s'(\omega_{\pm m})/(2\epsilon_0c)=\alpha_0(1+\delta^2)^{1/2}[E_1C_m+E_2C_{m\pm1}]^{1/2}, \tag{61}$$

where

$$C_m=\frac{2-\delta_{om}}{1+\delta^2+(E_1^2+E_2^2)/E_0^2}\,\frac{[(1-a_0^2)^{1/2}-1]^m}{a_0^m(1-a_0^2)^{1/2}}, \tag{62}$$

$$a_0=\frac{2E_1E_2/E_0^2}{1+\delta^2+(E_1^2+E_2^2)/E_0^2}, \tag{63}$$

and

$$\delta=(\omega_0-\omega)/\gamma.$$

$\delta_{0m}=1$ if $m=0$ and zero otherwise.

The polarization (61) will give rise to fields at $\omega_{\pm m}$, and these fields will induce a saturated polarization given by

$$\omega P_s(\omega_{\pm m})/(2\epsilon_0c)=\alpha_0C_0E(\omega_{\pm m}), \tag{64}$$

$$\omega P_c(\omega_{\pm m})/(2\epsilon_0c)=-\alpha_0(\omega_0-\omega)C_0E(\omega_{\pm m})/\gamma. \tag{65}$$

These results, with (15) and (16) give

$$\frac{dE_1}{dz}=\alpha_0\left(E_1C_0+\frac{E_2C_1}{2}\right), \tag{66}$$

$$n(\omega_1)=1-\frac{\alpha_0c}{\omega_1}\frac{(\omega_0-\omega_1)}{\gamma}\left(C_0+\frac{E_2C_1}{2E_1}\right), \tag{67}$$

with similar expressions at $\omega_2$, and at $\omega_{\pm m}\neq\omega_1,\omega_2$

$$\frac{dE_{\pm m}}{dz}=\alpha_0\left[E_{\pm m}C_0+\frac{(1+\delta^2)^{1/2}}{2}(E_1C_m+E_2C_{m\pm1})\right], \tag{68}$$

$$n(\omega_{\pm m})=1-\frac{c\alpha_0}{\omega_{\pm m}}\frac{(\omega_0-\omega_{\pm m})}{\gamma}C_0, \tag{69}$$

where in (68) and (69) we have assumed that the induced waves never get out of phase with the polarization (61), since $\Delta\omega\ll\gamma_a,\gamma_b$.

The above results hold so long as $m\Delta\omega\ll\gamma_a,\gamma_b$ and the induced fields remain small compared to $E_0$. Some of the coefficients $C_m$ are plotted in Fig. 4 for $E_1=E_2=E$ and $\delta=0$. In this case, the polarization at $\omega_{\pm m}$ due to $E_1$ and $E_2$ is determined by the sum of two successive $C_m$, and this sum is given by the vertical distance between the corresponding curves, taking into account the log scale. Thus we can see directly from Fig. 4 how the gain at various frequencies changes with increasing field strength.

If $E_1/E_0$ and $E_2/E_0$ are small compared to unity, the above results can be expanded to give the linear and lowest order nonlinear results as before.[14] Another interesting limiting case is $E_1=E_2=E$ and $E^2/E_0^2\gg1$. We find

$$2C_0+C_1\simeq E_0^2/E^2$$

and for $m$, $m-1\neq0$,

$$C_m+C_{m\pm1}\simeq\pm(-1)^mE_0^2/E^2.$$

The polarization due to $E_1$ and $E_2$ at each frequency is thus of order $E_0^2/E^2\ll1$ compared to the linear polarization. This is the case of extreme saturation, with the PID driven essentially to zero.

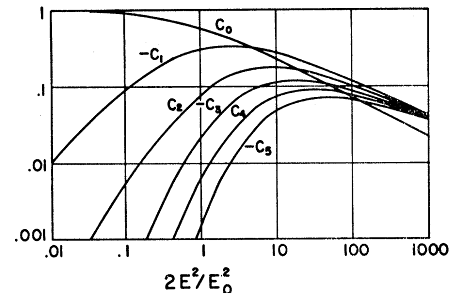


FIG. 4. Fourier gain modulation coefficients for the homogeneous line.

For the inhomogeneous line, $a[1+(E_1+E_2)^2/E_0^2]^{1/2}\ll1$, we find, analogous to (43) and (44),

$$\frac{\omega P(\mathbf{r},t)}{2\epsilon_0c}=\alpha\Bigg\{\Bigg[\exp(-x^2)\bigg(1+\frac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{E_0^2}\bigg)^{-1/2}-(2a/\pi^{1/2})(1-2xF(x))\Bigg][E_1\sin(\mathbf{k}_1\cdot\mathbf{r}-\omega_1t+\phi_1)$$
$$+E_2\sin(\mathbf{k}_2\cdot\mathbf{r}-\omega_2t+\phi_2)]-\Bigg[2F(x)/\pi^{1/2}-2a\bigg(1+\frac{E_1^2+E_2^2+2E_1E_2\cos\Delta_{12}}{E_0^2}\bigg)^{1/2}x\exp(-x^2)\Bigg]$$
$$\times[E_1\cos(\mathbf{k}_1\cdot\mathbf{r}-\omega_1t+\phi_1)+E_2\cos(\mathbf{k}_2\cdot\mathbf{r}-\omega_2t+\phi_2)]\Bigg\}. \quad (70)$$

Since for this case the cosine terms of (70) are not appreciably changed by saturation, we shall deal only with the sine terms and assume that the index of refraction is given by its linear value,

$$n(\omega)=1-(c\alpha/\omega)2F(x)/\pi^{1/2}, \quad (71)$$

where $x=(\omega_0-\omega)/(k_0u)$ as before. In the Appendix (70) is expanded into its Fourier components, with the result, using (15) and (16),

$$dE_1/dz=\alpha[\exp(-x^2)(E_1C_0'+E_2C_1'/2)-2aE_1(1-2xF(x))/\pi^{1/2}], \quad (72)$$

with a similar expression for $dE_2/dz$, and for the fields at $\omega_{\pm m}\neq\omega_1,\omega_2$,

$$dE_{\pm m}/dz=\alpha\{\exp(-x^2)(E_{\pm m}C_0'+E_1C_m'/2+E_2C_{m\pm1}/2)-2aE_{\pm m}[1-2xF(x)]/\pi^{1/2}\}, \quad (73)$$

where again we have assumed that the $E_{\pm m}$ fields remain in phase with their driving polarizations, and the $C_m'$ are given in the Appendix.

Some of the $C_m'$ coefficients are plotted in Fig. 5 for $E_1=E_2=E$, along with the corresponding $C_m$ coefficients from Fig. 3 for comparison. The induced gains can be seen to be very similar, within a factor $a\pi^{1/2}$. As before, these results hold for $m\Delta\omega\ll\gamma_a,\gamma_b$ and only as long as none of the induced field become appreciable compared to $E_0$. Here, we have the added restriction $a[1+(E_1^2+E_2^2)/E_0^2]^{1/2}\ll1$.

We can obtain an estimate of the induced fields expected by fixing the coefficients at their $z=0$ values. Thus for $2E^2/E_0^2=5$ we find from Fig. 4

$$C_0'\simeq0.5;\quad C_1'\simeq-0.3;\quad C_3'\simeq-0.05;\quad C_4'\simeq0.03.$$

If we take a moderate value of $\alpha=2$/m and an amplifier $\frac{1}{2}$m long, we find at line center $E_{+1}\approx0.2E$; $E_{+2}\approx0.05E$; $E_{+3}\approx0.01E$. For the 3.39-$\mu$ line with a saturation power of about 2 mW/cm², (Ref. 14) this means that for 6 mW/cm² input at $\omega_1$ and $\omega_2$, we would expect an output of roughly 0.2 mW/cm² at $2\omega_1-\omega_2$; 20 $\mu$W/cm² at

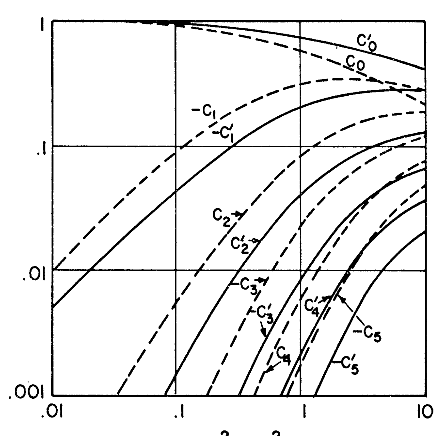


FIG. 5. Fourier gain modulation coefficients for the inhomogeneous line.

$3\omega_1-2\omega_2$. It should be emphasized that these fields are all produced directly by the interaction of the fields at $\omega_1$ and $\omega_2$ with the medium, and not by an iterative process, which would give much smaller higher order fields.

### B. More Than Two Input Components

For more than two incident fields with $|\omega_i-\omega_j| \ll\gamma_a,\gamma_b$ we can generalize the above results to find the polarization

$$\frac{\omega P(\mathbf{r},t)}{2\epsilon_0 c}=\frac{\alpha}{\pi}\sum_i\int_{-\infty}^{\infty}\frac{\exp(-y^2)dy[E_i a\sin(\mathbf{k}_i\cdot\mathbf{r}-\omega_i t+\phi_i)-E_i(x_i+y)\cos(\mathbf{k}_i\cdot\mathbf{r}-\omega_i t+\phi_i)]}{a^2[1+\sum_j E_j^2/E_0^2+\sum_{j\neq k}E_jE_k/E_0^2\cos\Delta_{jk}]+(x_i+y)^2}. \tag{74}$$

We see that (74) contains the saturation effects of all waves, with the PID being modulated at all possible combinations of the difference frequencies $\omega_i-\omega_j$. If there are a large number of waves within a frequency range $\gamma$ which have random phases and nearly equal amplitudes, the $\cos\Delta_{jk}$ summation will give zero and (74) gives

$$\frac{dE_i}{dz}=\frac{\alpha E_i}{(1+\sum_j E_j^2/E_0^2)^{1/2}}\mathrm{Re}w(x_i+ib), \tag{75}$$

where $b=a(1+\sum_j E_j^2/E_0^2)^{1/2}$, or in terms of the relative intensities $I_i=E_i^2/E_0^2$

$$\frac{dI_i}{dz}=\frac{2\alpha I_i}{(1+\sum_j I_j)^{1/2}}\mathrm{Re}w(x+ib), \tag{76}$$

where we have taken an average value $x$ for the frequency, which is essentially constant over the range of interest. Summing over all $i$ we find

$$\frac{dI_{\text{total}}}{dz}=\frac{2\alpha I_{\text{total}}}{(1+I_{\text{total}})^{1/2}}\mathrm{Re}w(x+ib). \tag{77}$$

Equation (77) is the same as the monochromatic field result (41). We can thus approximately describe the saturated amplification of a relatively broad, incoherent input, in particular the "super-radiant" emission of a high-gain amplifier. Thus the amplifier measurements of Gordon, White, and Rigden[15] using a super-radiant source, should approximately give the parameters for a single-frequency amplifier.

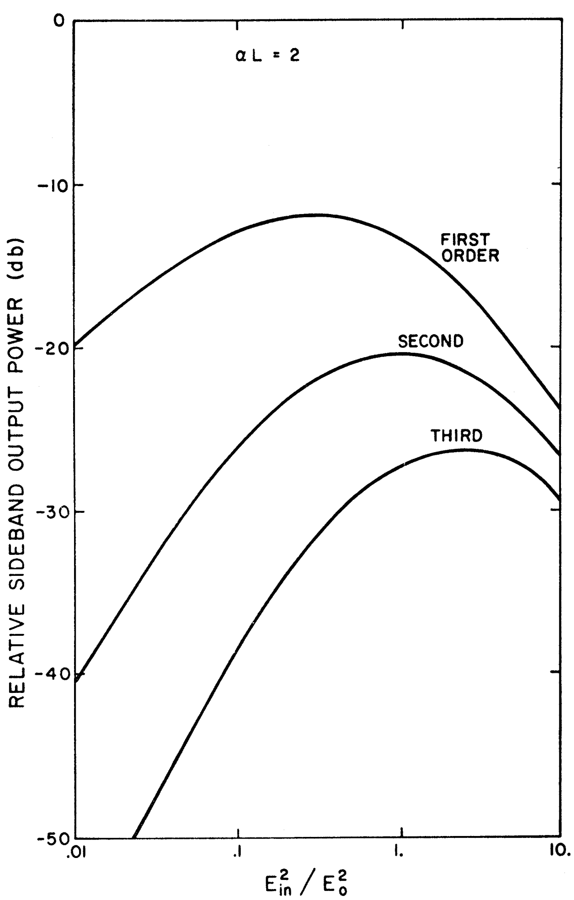


Fig. 6. Relative output sideband power as a function of field strength.

### C. Numerical Results for Two Components

The differential equations (66), (68), (72), (73), for the input and sideband fields were numerically integrated to obtain the relative sideband powers at the amplifier output. As an example, Fig. 6 shows the relative powers for the first three sidebands for $\alpha L=2$ and $\delta=0$ for a homogeneous line, as a function of the input power $E_1^2/E_0^2=E_2^2/E_0^2$. The computer integrations show that the relative sideband power at a fixed substantial input, e.g., $E_1^2/E_0^2=E_2^2/E_0^2=1$ increases approximately as $(\alpha L)^2$, neglecting the saturation due to the sideband fields, where $L$ is the amplifier length.

## VI. THE RUBY LASER; FREQUENCY DEPENDENCE OF THE POLARIZATION COEFFICIENTS

The ruby laser[21,22] can be treated in the above model if we assume that the lower (ground) level is pumped directly into the upper level with a "pumping time" $\tau_p$, i.e., neglecting the fast transition from the pump bands.

[21] T. H. Maiman, Nature 187, 493 (1960).

[22] R. J. Collins, D. F. Nelson, A. L. Schawlow, W. Bond, C. G. B. Garrett, and W. Kaiser, Phys. Rev. Letters 5, 303 (1960).

The required changes are

$$\begin{aligned}\lambda_a &\to \rho_{bb}(\mathbf{r},t)/\tau_p,\\ \gamma_a &\to 1/T_1,\\ \lambda_b &\to \rho_{aa}(\mathbf{r},t)/T_1,\\ \gamma_b &\to 1/\tau_p,\\ \gamma &\to 1/T_2,\end{aligned} \tag{78}$$

where $T_1$ is the fluorescence decay time and $T_2$ is the thermally shortened cross-relaxation time. A population inversion is obtained whenever $\tau_p < T_1$. For steady-state conditions we will have $\tau_p \approx T_1$, since

$$N_0 = N(1-\tau_p/T_1)/(1+\tau_p/T_1), \tag{79}$$

where $N$ is the density of chromium atoms.

The integral equation (28) becomes

$$N(r,t) = \int_{-\infty}^{t} dt_0 \exp(-(t-t_0)/T_1)\rho_{bb}(r,t_0)/\tau_p - \int_{-\infty}^{t} dt_0 \exp(-(t-t_0)/\tau_p)\rho_{aa}(\mathbf{r},t_0)/T_1$$
$$-\left(\frac{\omega_0}{2\hbar}\right)^2 \int_0^\infty dt_1 \int_0^\infty dt_2 (\exp(-t_1/T_1)+\exp(-t_1/\tau_p)) N(\mathbf{r},t-t_1-t_2) \sum_{\omega,\omega'} (\mathbf{P}_0\cdot\mathbf{E}_\omega)(\mathbf{P}_0\cdot\mathbf{E}_{\omega'})/(\omega\omega')$$
$$\times\{\exp[(i\Delta+i(\omega-\omega')t_1-[1/T_2-i(\omega_0-\omega')]t_2)]+\text{c.c.}\}, \tag{80}$$

where $N(\mathbf{r},t) = \rho_{aa}(\mathbf{r},t) - \rho_{bb}(\mathbf{r},t)$ as before. The $t_0$ integrals make (80) rather cumbersome. In order to study the same effects as previously, we will neglect the small effect of modulation on these excitation terms and replace them by a steady-state inversion. We note that in the zero-field case these terms return the $N_0$ of (79), and for $T_1 = \tau_p$ they return the saturated value, $C_0N_0$. With this in mind, we replace these terms by $N_0$. This neglect of modulation in the excitation process was implicitly done in the previous cases as well.

We are therefore left with the same integral equation as before. Since $T_1$ is a few milliseconds for ruby, the condition $\Delta\omega T_1 \ll 1$ previously used in studying field interactions is now difficult to realize in practice. For this case (and for the previous cases) we would like to study the solutions for arbitrary $\Delta\omega$. In order to do this, we recall that for two incident fields, $N(\mathbf{r},t)$ contains all harmonics of $\Delta\omega$, i.e., is of the form

$$N(\mathbf{r},t) = N_0 \textstyle\sum_p C_p \cos p\Delta + S_p \sin p\Delta, \tag{81}$$

where $\Delta = (\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r} - (\omega_1-\omega_2)t + \phi_1 - \phi_2$. If we substitute (81) into both sides of the integral equation and equate coefficients of $C_p$ and $S_p$ on both sides, we obtain the set of linear equations

$$C_0 = 1 - \left[\frac{E_1^2+E_2^2}{E_0^2}C_0 + \frac{E_1E_2}{E_0^2}C_1\right],$$

$$C_1 = -\left[\frac{E_1^2+E_2^2}{E_0^2}\frac{C_1+S_1\Delta\omega T_1}{1+(\Delta\omega T_1)^2} + \frac{E_1E_2}{E_0^2}\frac{2C_0+C_2+\Delta\omega T_1 S_2}{1+(\Delta\omega T_1)^2}\right],$$

$$C_j = -\left[\frac{E_1^2+E_2^2}{E_0^2}\frac{C_j+j\Delta\omega T_1 S_j}{1+(j\Delta\omega T_1)^2} + \frac{E_1E_2}{E_0^2}\frac{(C_{j-1}+C_{j+1})+j\Delta\omega T_1(S_{j-1}+S_{j+1})}{1+(j\Delta\omega T_1)^2}\right],$$

$$S_1 = -\left[\frac{E_1^2+E_2^2}{E_0^2}\frac{S_1-\Delta\omega T_1 C_1}{1+(\Delta\omega T_1)^2} + \frac{E_1E_2}{E_0^2}\frac{S_2-\Delta\omega T_1(2C_0+C_2)}{1+(\Delta\omega T_1)^2}\right],$$

$$S_j = -\left[\frac{E_1^2+E_2^2}{E_0^2}\frac{S_j-j\Delta\omega T_1 C_j}{1+(j\Delta\omega T_1)^2} + \frac{E_1E_2}{E_0^2}\frac{(S_{j-1}+S_{j+1})-j\Delta\omega T_1(C_{j-1}+C_{j+1})}{1+(j\Delta\omega T_1)^2}\right], \tag{82}$$

for $j \geq 2$, where

$$E_0^2 = \hbar^2/(T_1T_2P_0^2) \tag{83}$$

and we assume $(\omega_0-\omega_{1,2})T_2 \ll 1$, i.e., that we remain near line center. This infinite set of linear, algebraic equations can be solved for part of the coefficients provided the set of equations can be truncated, and this is possible if the values of the coefficients drop off rapidly enough for higher orders. This will certainly happen if $(E_1^2+E_2^2)/E_0^2$, the saturation parameter, is small compared to one.

It is evident that the set can also be truncated if $\Delta\omega T_1$ is large enough. For the case

$$(E_1^2+E_2^2)/\Delta\omega T_1 E_0^2 \ll 1 \tag{84}$$

the set can be truncated after the first order, giving

$$\begin{aligned}C_0 &= [1+(E_1^2+E_2^2)/E_0^2]^{-1},\\ C_1 &= -2E_1E_2/(E_0\Delta\omega T_1)^2,\\ S_1 &= 2E_1E_2/(E_0^2\Delta\omega T_1[1+(E_1^2+E_2^2)/E_0^2]),\end{aligned} \tag{85}$$

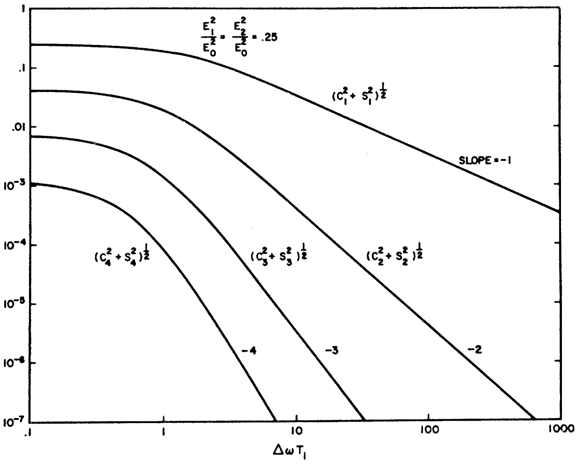


FIG. 7. Fourier gain modulation coefficients as a function of frequency spacing of inducing fields.

with higher order coefficients negligible compared with $S_1$.

A set of nine equations, i.e., terminated at the fourth order, was solved by computer for different values of the saturation parameter and $\Delta\omega T_1$; the resulting solutions were good for sufficiently small $(E_1^2+E_2^2)/E_0^2$ and up to $(E_1^2+E_2^2)/(\Delta\omega T_1 E_0^2)$ equal to about unity, for $E_1=E_2$. The dependence of the net polarization coefficients $(C_j^2+S_j^2)^{1/2}$ on $\Delta\omega$ is shown in Fig. 7 for $E_1^2/E_0^2 = E_2^2/E_0^2=0.25$. Note that the slope of the falloff for large $\Delta\omega T_1$ is equal to the sideband order. For a fixed $\Delta\omega T_1$, the coefficients increase with increasing $E_1$ and $E_2$, until $(E_1^2+E_2^2)/E_0^2 \approx \Delta\omega T_1$, after which they probably behave similarly to the large field limit for small $\Delta\omega$, i.e., all becoming smaller but more nearly equal. Unfortunately, for this range of parameters the set of equations cannot be truncated at a reasonably low order.

These results on the frequency dependence of the polarization coefficients should apply qualitatively to the earlier situations studied, taking into account the unequal decay rates $\gamma_a$ and $\gamma_b$ of the two laser levels, and the smaller relative linewidth $\gamma$.

## VII. CONCLUSION AND DISCUSSION

Following Lamb's model, an analysis of strong-field effects in laser media was made. In particular, the "intermodulation" effect was considered, and interpreted as a time-dependent saturation leading to parametric gain modulation. The frequency dependence of the interaction was studied for a typical solid-state laser.

Schulz-DuBois[23] and Tabor, Chen and Schulz-DuBois[24] previously studied the same effect for microwave masers, using a perturbation approach, with the conclusion that the effect was extremely small and required pulsed operation to be observed at their experimental $\Delta\omega/2\pi$ of 30 Mc/sec. Since for this case $\Delta\omega T_1 \approx 10^7$ (where $T_1$ is the spin-lattice relaxation time) and $P/P_{\text{sat}} \lesssim 10^4$, the effect should indeed be small, and the perturbation analysis[23] should be completely adequate. However, the conclusion that the short spin-spin relaxation time $T_2$ causes the small magnitude of the intermodulation effect in the ruby maser does not seem to be accurate, except to the extent that a small $T_2$ gives a large saturation power. For a given power, the amount of gain modulation is determined by how well the populations can follow the beating fields, i.e., by $T_1$, while $T_2$ determines the linewidth (and, with $T_1$, the saturation power). The maser results were determined primarily by the large $\Delta\omega T_1$; a smaller $\Delta\omega$ would have correspondingly increased the effect, as was indicated elsewhere in the discussion.[23]

A small $T_1$ occurs for most solid state lasers (e.g., for ruby $T_1 \approx 3\times10^{-3}$ sec), so that again a large $\Delta\omega T_1$ will be the rule. Again it is possible to use large signals and pulsed operation[23,24] to obtain a situation where fairly large amounts of intermodulation could be observed. Thus in a giant-pulse laser one typically has $\Delta\omega T_1 = 4\times10^6$ and $P/P_{\text{sat}}=10^7$, a case where a perturbation analysis would not be adequate.

Preliminary experimental results[13] with the helium-neon gas laser at 3.39 $\mu$ are in good agreement with the above theory.

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## APPENDIX: EVALUATION OF THE FOURIER COEFFICIENTS

For the homogeneous coefficients we have

$$C_m = \frac{(2-\delta_{om})/\pi}{1+\delta^2+(E_1^2+E_2^2)/E_0^2} \int_0^\pi \frac{\cos mu\,du}{1+a_0 \cos u}, \quad \text{(A1)}$$

where

$$a_0 = \frac{2E_1E_2/E_0^2}{1+\delta^2+(E_1^2+E_2^2)/E_0^2}. \quad \text{(A2)}$$

The integral in (A1) is tabulated,[25] giving

$$C_m = \frac{(-1)^m(2-\delta_{om})}{1+\delta^2+(E_1^2+E_2^2)/E_0^2} \frac{[1-(1-a_0^2)^{1/2}]^m}{a_0^m(1-a_0^2)^{1/2}}. \quad \text{(A3)}$$

[23] E. O. Schulz-DuBois, Proc. IEEE **52**, 644 (1964).
[24] W. J. Tabor, F. S. Chen, and E. O. Schulz-DuBois, Proc. IEEE **52**, 656 (1964).
[25] H. B. Dwight, *Tables of Integrals and Other Mathematical Data* (MacMillan Publishers, New York, 1961), p. 219, No. 858.536.

For the inhomogeneous coefficients, we have

$$C_m'=\frac{(2-\delta_{om})/\pi}{[1+(E_1^2+E_2^2)/E_0^2]^{1/2}}\int_0^\pi \frac{\cos mu\,du}{(1+a_0'\cos u)^{1/2}}, \quad \text{(A4)}$$

where

$$a_0'=\frac{2E_1E_2/E_0^2}{1+(E_1^2+E_2^2)/E_0^2}. \quad \text{(A5)}$$

With the substitution $V=\exp(iu)$, the integral in (A4) becomes

$$\mathrm{Re}\left(\frac{2}{a_0'}\right)^{1/2}\frac{1}{i}\int_c \frac{V^{m-1/2}dV}{(V-V_1)^{1/2}(V-V_2)^{1/2}}, \quad \text{(A6)}$$

where

$$\begin{aligned} -V_1&=(1+(1-a_0'^2)^{1/2}/a_0', \\ -V_2&=(1-(1-a_0'^2)^{1/2}/a_0', \end{aligned} \quad \text{(A7)}$$

and the contour $c$ is the upper half unit circle in the complex $V$ plane. Evaluating (A6) by standard methods gives

$$C_m'=\frac{(2-\delta_{om})(-1)^m(8/a_0')^{1/2}}{\pi[1+(E_1^2+E_2^2)/E_0^2]^{1/2}}\int_0^d \frac{(d^2-t^2)^{m-1/2}dt}{(c^2+t^2)^{1/2}}, \quad \text{(A8)}$$

where $d^2=-V_1$, $c^2=V_1-V_2$ are greater than zero. Equation (A8) can be used to explicitly evaluate the $C_m'$. An alternative expression is obtained by expanding $(1+a_0'\cos u)^{-1/2}$ by the binomial theorem, where the coefficient of $(\cos u)^m$ is

$$\frac{(-1)^m(2m-1)!!a_0'^m}{2^m m!}. \quad \text{(A9)}$$

The coefficient of $\cos(m-2p)u$ in the expansion of $(\cos u)^m$ is [26]

$$2^{-m+1}C_p^m, \quad \text{(A10)}$$

except for $p=m/2$ an integer, when it is

$$2^{-m}C_p^m, \quad \text{(A11)}$$

where the $C_p^m$ are the binomial coefficients. Combining these coefficients, we find an infinite series for the $C_m'$,

$$C_m'=\frac{(2-\delta_{om})(-a_0'/4)^m}{[1+(E_1^2+E_2^2)/E_0^2]^{1/2}} \times\sum_{p=0}^{\infty}\left(\frac{a_0'^2}{16}\right)^p\frac{(4p+2m-1)!!}{p!(m+p)!}, \quad \text{(A12)}$$

which converges absolutely for $a_0'<1$. In (A12) we define $(4p+2m-1)!!=1$ for $m=p=0$.

[26] Reference 25, p. 82, No. 404.