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Citation for published version (APA):

Lamb, W. E. (1964). Theory of an optical maser. *Physical Review*, 134, A1429-A1450.

Document status and date:

Published: 01/01/1964

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

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Theory of an Optical Maser*

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(Received 13 January 1964)

A theoretical model for the behavior of an optical maser is presented in which the electromagnetic field is treated classically, and the active medium is made up of thermally moving atoms which acquire nonlinear electric dipole moments under the action of the field according to the laws of quantum mechanics. The corresponding macroscopic electric polarization of the medium acts as a source for an electromagnetic field. The self-consistency requirement that a quasistationary field should be sustained by the induced polarization leads to equations which determine the amplitudes and frequencies of multimode oscillation as functions of the various parameters characterizing the maser. Among the results obtained are: threshold conditions, single-mode output as a function of cavity tuning, frequency pulling and pushing, mode competition phenomena including frequency locking, production of combination tones, and population pulsations. A more approximate discussion of maser action using rate equations is also given in which the concept of "hole burning" plays a role.

1. INTRODUCTION

THIS paper gives a theoretical description of the operation of multimode maser oscillators. The type of approach is particularly suitable for gaseous optical masers of the type suggested by Schawlow and Townes,¹ and first realized experimentally by Javan, Bennett, and Herriott,² but the equations should also find use in the description of some features of solid-state optical masers.

2. BASIS FOR CALCULATION

We consider a high- Q multimode cavity in which there is a given *classical* electromagnetic field acting on a material medium which consists of a collection of atoms described by the laws of quantum mechanics. No attempt is made to consider noise due to spontaneous emission and thermal, density, or quantum fluctuations. The high degree of spectral purity observed by Javan and co-workers³ suggests that these should be good approximations.

The effect of the electromagnetic field on the atoms in the cavity is to produce a macroscopic electric polarization $\mathbf{P}(\mathbf{r}, t)$ of the medium. This acts as a source for the electromagnetic field in accordance with Maxwell's equations. The conditions for self-consistency (that the field produced should be equal to the field assumed) determine the amplitudes and frequencies of the possible oscillations. The calculations will include nonlinear effects, so that phenomena of frequency pulling and pushing, mode competition, frequency locking, etc., can be described.

The thermal motion of an atom during its natural decay time may carry it several wavelengths through the standing wave pattern of the electromagnetic field. As a result, the atom "sees" Doppler-shifted optical frequencies which depend on its trajectory. This important circumstance considerably influences the behavior of the Javan-Bennett-Herriott maser. When, however, thermal motion is neglected the equations of the paper can be used in a model calculation for an ideal solid-state optical maser.

We will assume that only two atomic states a and b contribute to the maser action. As a related simplification the vector character (polarization) of the electromagnetic field will be ignored. In order to ensure that our analysis should apply, it would be desirable to have the optical configuration favor one plane of polarization, as with windows of the Brewster's angle type. The more complicated problem of a general state of polarization will be dealt with in another paper.

A cavity of the Fabry-Perot type used by Javan, Bennett, and Herriott has, of course, a continuum of modes because it is not enclosed by reflecting walls. However, it follows from work of Fox and Li⁴ that there are discrete sets of quasimodes for which the diffractive leakage from the tube is small. The cavity modes of highest Q are the even symmetric ones whose circular frequencies are given by

$$\Omega_n = \pi n c / L, \quad (1)$$

where c is the velocity of light, L is the distance between the reflecting plates ($L \sim 100$ cm) and n is a large integer, typically of order 2×10^6 . Fox and Li have shown that the modes of next highest Q are those possessing odd radial symmetry, which, for typical geometry differ by about 1 Mc/sec from the former modes. Our discussion will be specifically, but not inevitably, aimed at the modes of highest Q .

* This work was supported in part by the U. S. Air Force Office of Scientific Research. The main results of the paper were reported at the Third International Conference on Quantum Electronics, Paris, February, 1963. Lectures on some of the material were given at the 1963 Varenna Summer School.

¹ A. L. Schawlow and C. H. Townes, *Phys. Rev.* **112**, 1940 (1958).

² A. Javan, W. R. Bennett and D. R. Herriott, *Phys. Rev. Letters* **6**, 106 (1961).

³ T. S. Jaseja, A. Javan, and C. H. Townes, *Phys. Rev. Letters* **10**, 165 (1963).

⁴ A. G. Fox and T. Li, *Bell System Tech. J.* **40**, 61 (1961).

3. ELECTROMAGNETIC FIELD EQUATIONS

We write Maxwell's equations in mks units as

$$\begin{aligned} \operatorname{div} \mathbf{D} &= 0 & \operatorname{curl} \mathbf{E} &= -\partial \mathbf{B} / \partial t \\ \operatorname{div} \mathbf{B} &= 0 & \operatorname{curl} \mathbf{H} &= \mathbf{J} + \partial \mathbf{D} / \partial t, \end{aligned} \quad (2)$$

where

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{D}, \quad \mathbf{B} = \mu_0 \mathbf{H}, \quad \mathbf{J} = \sigma \mathbf{E}. \quad (2a)$$

To an approximation whose validity will be discussed in another paper, the array of excited atoms may be regarded as a medium with an electrical state described by a macroscopic polarization $\mathbf{P}(\mathbf{r}, t)$ (electric dipole moment density). In order to avoid a complicated boundary value problem, it is convenient to assume the presence of a lossy medium with an Ohmic conductivity σ adjusted to give the desired damping of a normal mode. The electric field then obeys a wave equation

$$\operatorname{curl} \operatorname{curl} \mathbf{E} + \mu_0 \sigma \partial \mathbf{E} / \partial t + \mu_0 \epsilon_0 \partial^2 \mathbf{E} / \partial t^2 = -\mu_0 \partial^2 \mathbf{P} / \partial t^2. \quad (3)$$

In the subsequent calculations the main effect of the space dependence of $\mathbf{E}(x, y, z, t)$ comes from the motion of the excited atoms through the field which leads to amplitude modulation of the fields seen by the atoms. The analysis of Fox and Li for the even symmetric modes indicates that the electric field does not vary rapidly across the tube diameter. Accordingly we take only the axial variation of \mathbf{E} into account. Then $\operatorname{curl} \operatorname{curl} \mathbf{E}$ is replaced by $-\partial^2 E / \partial z^2$, where z is the axial coordinate, and E is the transverse electric field. For the n th normal mode (unnormalized), we have eigenfunctions

$$U_n(z) = \sin K_n z, \quad (4)$$

with wave number

$$K_n = n\pi / L, \quad (5)$$

where n is a large integer.

In the presence of a given polarization $P(z, t)$, quasi-stationary forced oscillations of the electric field can be expanded in normal mode eigenfunctions

$$E(z, t) = \sum_n A_n(t) U_n(z), \quad (6)$$

where the amplitudes $A_n(t)$ obey a differential equation of a forced, damped simple harmonic oscillator

$$\frac{d^2 A_n}{dt^2} + \left(\frac{\sigma}{\epsilon_0} \right) \frac{dA_n}{dt} + \Omega_n^2 A_n = - \left(\frac{1}{\epsilon_0} \right) \frac{d^2 P_n(t)}{dt^2}, \quad (7)$$

in which $P_n(t)$ is the space Fourier component of $P(z, t)$

$$P_n(t) = \frac{2}{L} \int_0^L dz P(z, t) \sin K_n z. \quad (8)$$

Since $P_n(t)$ will be very nearly monochromatic at an optical frequency⁵ (o.f.) ν , we replace its second time

⁵ We adopt the convention that all symbols for frequencies should denote circular frequencies. A numerical value, e.g., 150 Mc/sec, however, denotes an ordinary frequency. A decay constant like γ_α which denotes a reciprocal life time $1/\tau_\alpha$ often plays the role of a circular frequency. Numerical values of γ_α will be given as ordinary frequencies.

derivative by $-\nu^2 P_n$ on the right side of Eq. (7). Adjusting the fictional conductivity σ to give the desired Q_n of the n th mode, we write

$$\sigma = \epsilon_0 \nu / Q_n. \quad (9)$$

Then $A_n(t)$ obeys

$$\frac{d^2 A_n}{dt^2} + \left(\frac{\nu}{Q_n} \right) \frac{dA_n}{dt} + \Omega_n^2 A_n = \left(\frac{\nu^2}{\epsilon_0} \right) P_n. \quad (10)$$

In the typical gaseous optical maser, the separation of the principal modes $\Delta \sim 150$ Mc/sec is much larger than the cavity mode band width $\nu/Q \sim 1$ Mc/sec. Hence we may hope to neglect time Fourier components of $A_n(t)$ and $P_n(t)$ which are at frequencies far from the cavity resonance frequency Ω_n , and write⁶

$$A_n(t) = E_n(t) \cos(\nu_n t + \varphi_n(t)), \quad (11)$$

and

$$\begin{aligned} P_n(t) &= C_n(t) \cos(\nu_n t + \varphi_n(t)) \\ &\quad + S_n(t) \sin(\nu_n t + \varphi_n(t)), \end{aligned} \quad (12)$$

where the amplitudes $E_n(t)$ and phases $\varphi_n(t)$, as well as the in-phase and quadrature coefficients $C_n(t)$ and $S_n(t)$ are slowly varying functions of t which, together with the frequencies ν_n , are still to be determined. The expressions (11) and (12) are put into Eq. (10) with only the first time derivatives of $E_n(t)$ and $\varphi_n(t)$ retained. Equating the coefficients of $\cos(\nu_n t + \varphi_n)$ and $\sin(\nu_n t + \varphi_n)$ separately to zero, and further neglecting small terms involving $\nu_n \dot{E}_n / Q_n$, $\dot{\varphi}_n \dot{E}_n$ and $\nu_n \dot{\varphi}_n E_n / Q_n$, and recognizing that $\nu_n + \dot{\varphi}_n$ is very close to Ω_n , we find the self-consistency equations

$$(\nu_n + \dot{\varphi}_n - \Omega_n) E_n = -\frac{1}{2} (\nu / \epsilon_0) C_n \quad (13)$$

and

$$\dot{E}_n + \frac{1}{2} (\nu / Q_n) E_n = -\frac{1}{2} (\nu / \epsilon_0) S_n, \quad (14)$$

which serve to determine the amplitudes, frequencies and phases of the o.f. radiation once the polarization state of the medium is known in terms of the $E_n(t)$.

4. POLARIZATION OF THE MEDIUM

The maser action arises from the establishment of a negative temperature distribution for the two excited states a and b of the atoms constituting the medium as shown in Fig. 1. The ground state, far below a and b , is not shown. Consider what happens to an atom which at time t_0 is excited by some process (electron bombardment, collision of the second kind, absorption of resonance radiation, decay from some higher excited

⁶ The representation of an arbitrary function $A_n(t)$ in the form (11) in terms of a variable amplitude $E_n(t)$ and phase $\varphi_n(t)$ is not unique. Despite this, because of the use of the rotating wave approximation it seems possible through Eqs. (13) and (14) to determine both amplitude and phase. [The positive frequency part of (11) is a complex function very closely equal to $A_n^{(+)}(t) = E_n(t) \exp -i(\nu_n t + \varphi_n(t))$ which does have a unique amplitude and phase.]

state, etc.) into the upper maser state a . Let the atom be at position \mathbf{r}_0 at t_0 , and have velocity \mathbf{v} . For the present, we neglect collisions, so that at time $t > t_0$ the atom will be at $\mathbf{r} = \mathbf{r}_0 + \mathbf{v}(t - t_0)$. If there is an o.f. electric field $\mathbf{E}(\mathbf{r}, t)$ in the cavity, the atom sees a time-dependent field $E(\mathbf{r}_0 + \mathbf{v}t - \mathbf{v}t_0, t)$ for $t > t_0$. Associated with this field is a time-dependent perturbation energy whose matrix element is

$$\hbar V(t) = -\wp E(\mathbf{r}_0 + \mathbf{v}t - \mathbf{v}t_0, t), \quad (15)$$

where \wp (assumed real) is the matrix element for the electric dipole moment of the atom between states a and b . The perturbation causes the atomic wave function to become a time-dependent linear combination $a(t)\psi_a + b(t)\psi_b$. The quantum-mechanical average value of the electric dipole operator for the atom is $(a^*b + ab^*)\wp$.

To follow the time-dependent wave function (in the subspace of ψ_a and ψ_b), we start from the equations of time-dependent perturbation theory

$$\begin{aligned} i\dot{a} &= W_a a + V(t)b - \frac{1}{2}i\gamma_a a, \\ i\dot{b} &= W_b b + V(t)a - \frac{1}{2}i\gamma_b b, \end{aligned} \quad (16)$$

in which the radiative decay of states a and b is described by phenomenological terms containing the decay constants γ_a and γ_b for the two states. Here $\hbar W_a$ and $\hbar W_b$ are the unperturbed energies of states a and b , and the matrix element of the perturbation $V(t)$ is given by Eq. (15).

If the motion of the atom were neglected, and if the maser were working in a single cavity mode, $V(t)$ would be monochromatic, and the rotating wave approximation would allow the Eqs. (16) to be integrated exactly. Even so, there are great algebraic simplifications to be gained by going over to a density matrix description⁷ of an ensemble of atoms consisting of all those of a given category which are produced during all times $t_0 < t$. A theory of maser action in this case has already been given⁸ which is valid when the signals are strong enough to fully saturate the transition $a \leftrightarrow b$. For multimode operation, such an exact solution can no longer be obtained. However, the simpler theory can help with the interpretation of our rather complicated equations, and it will be discussed in Secs. 16–20.

When atomic motion through the electromagnetic field is taken into account an atom does not see a monochromatic perturbation even in single-mode operation. The equations can only be solved in a perturbation expansion of the solution in powers of the $E_n(t)$. It is still advantageous to use the density matrix method, considering first only those atoms characterized by

⁷ W. E. Lamb, Jr. and T. M. Sanders, Jr., *Phys. Rev.* **119**, 1901 (1960), especially pp. 1902–1903; L. R. Wilcox and W. E. Lamb, Jr., *ibid.* **119**, 1915 (1960), especially p. 1928.

⁸ W. E. Lamb, Jr., *Quantum Mechanical Amplifiers, in Lectures in Theoretical Physics*, edited by W. E. Brittin and B. W. Downs (Interscience Publishers, Inc., New York, 1960), Vol. II, especially pp. 472–476.

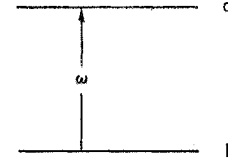


FIG. 1. Two excited energy levels a and b between which the maser action takes place. The levels have a resonance transition frequency $\omega > 0$, and are given phenomenological decay constants γ_a and γ_b . The excitation of the states is described by the functions $\lambda_\alpha(\mathbf{r}_0, t_0, \mathbf{v})$ which are introduced in Eq. (22).

a , \mathbf{r}_0 , t_0 , \mathbf{v} . The density matrix

$$\begin{aligned} \rho(a, \mathbf{r}_0, t_0, \mathbf{v}, t) &= \begin{pmatrix} |a|^2 & ab^* \\ a^*b & |b|^2 \end{pmatrix} \\ &= \begin{pmatrix} \rho_{aa} & \rho_{ab} \\ \rho_{ba} & \rho_{bb} \end{pmatrix} \end{aligned} \quad (17)$$

obeys an equation of motion

$$\dot{\rho} = -i[\mathcal{H}, \rho] - \frac{1}{2}(\Gamma\rho + \rho\Gamma), \quad (18)$$

where Γ is the diagonal matrix

$$\Gamma = \begin{pmatrix} \gamma_a & 0 \\ 0 & \gamma_b \end{pmatrix} \quad (19)$$

and the Hamiltonian matrix \mathcal{H} is

$$\mathcal{H} = \begin{pmatrix} W_a & V(t) \\ V(t) & W_b \end{pmatrix}, \quad (20)$$

with $V(t)$, as given by Eq. (15), having a complicated time dependence because of atomic motion. A solution of Eq. (18) which satisfies the initial conditions

$$\rho(a, \mathbf{r}_0, t_0, \mathbf{v}, t_0) = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \quad (21)$$

is required. The average electric dipole moment corresponding to this density matrix ρ is $\wp(\rho_{ab} + \rho_{ba})$.

To obtain the macroscopic polarization $P(\mathbf{r}, t)$ we have to combine the contributions of all atoms which arrive at \mathbf{r} at time t , no matter when or where they were excited to state a , and also a similar contribution from atoms excited initially to state b . Let $\lambda_\alpha(\mathbf{r}_0, t_0, \mathbf{v})$ be the number of atoms excited to state $\alpha = a, b$ per unit time per unit volume. We have

$$\begin{aligned} P(\mathbf{r}, t) &= \wp \sum_{\alpha=a, b} \int_{-\infty}^t dt_0 \int d\mathbf{r}_0 \int d\mathbf{v} \lambda_\alpha(\mathbf{r}_0, t_0, \mathbf{v}) \\ &\quad \times [\rho_{ab}(\alpha, \mathbf{r}_0, t_0, \mathbf{v}, t) + \rho_{ba}(\alpha, \mathbf{r}_0, t_0, \mathbf{v}, t)] \\ &\quad \times \delta(\mathbf{r} - \mathbf{r}_0 - \mathbf{v}(t - t_0)). \end{aligned} \quad (22)$$

In practice, $\lambda_\alpha(\mathbf{r}_0, t_0, \mathbf{v})$ will be a slowly varying function

of \mathbf{r}_0 so that it can be replaced by $\lambda_\alpha(\mathbf{r}, t_0, \mathbf{v})$. After integration over \mathbf{r}_0

$$P(\mathbf{r}, t) = \wp \sum_{\alpha=a, b} \int_{-\infty}^t dt_0 \int d\mathbf{v} \quad (23)$$

$$\times \lambda_\alpha(\mathbf{r}, t_0, \mathbf{v}) [\rho_{ab}(\alpha, \mathbf{r} - \mathbf{v}(t - t_0), t_0, \mathbf{v}, t) + \text{conj.}].$$

Similarly, we will have use for a density matrix describing an ensemble of atoms which arrive at \mathbf{r} with velocity \mathbf{v} at time t regardless of their place \mathbf{r}_0 , time t_0 or state $\alpha = a, b$ of excitation. This will be denoted by

$$\rho(\mathbf{r}, \mathbf{v}, t) = \sum_{\alpha=a, b} \int_{-\infty}^t dt_0 \int d\mathbf{r}_0 \lambda_\alpha(\mathbf{r}_0, t_0, \mathbf{v}) \quad (24)$$

$$\times \rho(\alpha, \mathbf{r}_0, t_0, \mathbf{v}, t) \delta(\mathbf{r} - \mathbf{r}_0 - \mathbf{v}t + \mathbf{v}t_0).$$

The density matrix resulting from (24) by integration over all velocities will be denoted by $\rho(\mathbf{r}, t)$.

5. INTEGRATION OF THE EQUATIONS OF MOTION

The matrix equation of motion for the density matrix $\rho(\alpha, \mathbf{r}_0, t_0, \mathbf{v}, t)$ has components

$$\dot{\rho}_{ab} = -i\omega\rho_{ab} - \gamma_{ab}\rho_{ab} + iV(t)(\rho_{aa} - \rho_{bb}), \quad (25)$$

$$\dot{\rho}_{aa} = -\gamma_a\rho_{aa} + iV(t)(\rho_{ab} - \rho_{ba}),$$

$$\dot{\rho}_{bb} = -\gamma_b\rho_{bb} - iV(t)(\rho_{ab} - \rho_{ba}), \quad (26)$$

$$\rho_{ba} = \rho_{ab}^*,$$

where

$$\gamma_{ab} = \frac{1}{2}(\gamma_a + \gamma_b) \quad (26a)$$

and

$$\omega = W_a - W_b > 0. \quad (27)$$

We consider first the case of excitation to the upper maser state a . At $t = t_0$, $\rho_{aa} = 1$ and $\rho_{bb} = \rho_{ab} = \rho_{ba} = 0$. The solution to any desired order in the perturbation $V(t)$ can be obtained by iteration. There are contributions to $\rho_{ab} = \rho_{ba}^*$ in first and third order, to ρ_{bb} in second order, and to ρ_{aa} in zeroth and second order. Thus, in zeroth order,

$$\rho_{aa}^{(0)}(a, \mathbf{r}_0, t_0, \mathbf{v}, t) = \exp -\gamma_a(t - t_0) \quad (28)$$

and the first-order contribution to ρ_{ab} is

$$\rho_{ab}^{(1)}(a, \mathbf{r}_0, t_0, \mathbf{v}, t) = i \int_{t_0}^t dt' V(t') \exp[(\gamma_{ab} + i\omega)(t' - t) + \gamma_a(t_0 - t')], \quad (29)$$

while the third-order contribution is

$$\rho_{ab}^{(3)}(a, \mathbf{r}_0, t_0, \mathbf{v}, t) = i \int_{t_0}^t dt' V(t') \quad (30)$$

$$\times [\rho_{aa}^{(2)}(a, \mathbf{r}_0, t_0, \mathbf{v}, t') - \rho_{bb}^{(2)}(a, \mathbf{r}_0, t_0, \mathbf{v}, t')] \times \exp(\gamma_{ab} + i\omega)(t' - t),$$

where

$$\rho_{aa}^{(2)}(a, \mathbf{r}_0, t_0, \mathbf{v}, t') = - \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt''' V(t'') V(t''') \quad (31)$$

$$\times \{ \exp[\gamma_a(t'' - t') + (\gamma_{ab} + i\omega)(t''' - t'')] + \gamma_a(t_0 - t''') \} + \text{conj.},$$

and similarly

$$\rho_{bb}^{(2)}(a, \mathbf{r}_0, t_0, \mathbf{v}, t') = + \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt''' V(t'') V(t''') \quad (32)$$

$$\times \{ \exp[\gamma_b(t'' - t') + (\gamma_{ab} + i\omega)(t''' - t'')] + \gamma_a(t_0 - t''') \} + \text{conj.}.$$

6. FIRST-ORDER THEORY

In order to convert the expression (29) for $\rho_{ab}^{(1)}$ into a macroscopic polarization, we must first calculate

$$\rho_{ab}^{(1)}(a, \mathbf{r}, \mathbf{v}, t) = \int_{-\infty}^t dt_0 \rho_{ab}^{(1)}(a, \mathbf{r}_0 = \mathbf{r} - \mathbf{v}t + \mathbf{v}t_0, t_0, \mathbf{v}, t) \quad (33)$$

as in Eq. (24).

The perturbation $V(t')$ acting at time t' on the atom specified by $\mathbf{r}_0, t_0, \mathbf{v}$ is $-(\wp/\hbar)E(\mathbf{r}_0 + \mathbf{v}(t' - t_0), t')$, but for Eq. (33) we require this for an atom characterized by $\mathbf{r}_0 = \mathbf{r} - \mathbf{v}t + \mathbf{v}t_0, \mathbf{v}, t_0$, for which the effective perturbation

$$V(t') = -(\wp/\hbar)E(\mathbf{r} - \mathbf{v}(t - t'), t') \quad (34)$$

does not depend on t_0 . We may then perform the above integration over t_0 if we treat λ_a as a slowly varying function of t_0 and evaluate it at t . For the first-order terms, we have to deal with an expression of the form

$$\int_{-\infty}^t dt_0 \int_{t_0}^t dt' F(t, t') e^{\gamma_a(t_0 - t')}.$$

By an interchange⁹ of the order of integrations this becomes

$$\int_{-\infty}^t dt' \int_{-\infty}^{t'} dt_0 F(t, t') e^{\gamma_a(t_0 - t')} = (1/\gamma_a) \int_{-\infty}^t dt' F(t, t'). \quad (35)$$

Let us assume now that the maser oscillator is running simultaneously in M cavity modes, so that

$$E(\mathbf{r}, t) = \sum_{\mu=1}^M E_\mu(t) U_\mu(\mathbf{r}) \cos(\nu_\mu t + \varphi_\mu(t)), \quad (36)$$

⁹ In each double integral an integration is carried over the same triangular area in the t_0, t' plane.

where $E_\mu(t)$ and $\varphi_\mu(t)$ are slowly varying functions of time. We make a rotating wave approximation by keeping only exponential factors like $\exp i(\omega - \nu_\mu)t'$ and neglecting rapidly varying exponentials like $\exp i(\omega + \nu_\mu)t'$. Then $\rho_{ab}^{(1)}(a, \mathbf{r}, \mathbf{v}, t) =$

$$-\frac{1}{2}i\left(\frac{\wp}{\hbar}\right)\left[\frac{\lambda_a(\mathbf{r}, v, t)}{\gamma_a}\right]\sum_{\mu=1}^M\int_{-\infty}^t dt' E_\mu(t') \\ \times U_\mu(\mathbf{r} - \mathbf{v}(t-t')) \exp -i(\nu_\mu t + \varphi_\mu(t')) \\ \times \exp[-\gamma_{ab} + i(\nu_\mu - \omega)](t-t'). \quad (37)$$

We also assume that the amplitudes $E_\mu(t')$ and phases $\varphi_\mu(t')$ do not vary much in a time $1/\gamma_{ab}$, so that they can be evaluated at time t . With a change of variable of integration from t' to $\tau' = t - t'$, we find

$$\rho_{ab}^{(1)}(a, \mathbf{r}, \mathbf{v}, t) = \\ -\frac{1}{2}i\left(\frac{\wp}{\hbar}\right)\left[\frac{\lambda_a(\mathbf{r}, \mathbf{v}, t)}{\gamma_a}\right]\sum_{\mu=1}^M E_\mu(t) \exp -i(\nu_\mu t + \varphi_\mu(t)) \\ \times \int_0^\infty d\tau' U_\mu(\mathbf{r} - \mathbf{v}\tau') \exp -(\gamma_{ab} + i(\omega - \nu_\mu))\tau'. \quad (38)$$

The corresponding contribution to the polarization of the medium is

$$P^{(1)}(a, \mathbf{r}, \mathbf{v}, t) \\ = -\frac{1}{2}i(\wp^2/\hbar)\left[\lambda_a(\mathbf{r}, \mathbf{v}, t)/\gamma_a\right] \\ \times \sum_{\mu} E_\mu(t) \left\{ \exp -i(\nu_\mu t + \varphi_\mu) \int_0^\infty d\tau' U_\mu(\mathbf{r} - \mathbf{v}\tau') \right. \\ \left. \times \exp -(\gamma_{ab} + i(\omega - \nu_\mu))\tau' \right\} + \text{conj.} \quad (39)$$

Let us first assume that the excitation rate density has the form¹⁰

$$\lambda_\alpha(\mathbf{r}, \mathbf{v}, t) = W(\mathbf{v})\Lambda_\alpha(\mathbf{r}, t) \quad \alpha = a, b, \quad (40)$$

where $W(\mathbf{v})$ is the normalized velocity distribution function and $\Lambda_\alpha(\mathbf{r}, t)$ is the number of atoms excited to state α per unit volume and time. Because we are assuming a spatial dependence of the electric field only on z , we may change over from a three- to a one-dimensional description. Then the velocity distribution $W(v)$ refers to the z component v of \mathbf{v} , and \mathbf{r} is replaced by z .

It will be noted that the quantity $P(a, z, v, t)$ is proportional to $\Lambda_a(z, t)/\gamma_a$. When we now consider the contribution of atoms excited to the lower maser state b there is a complication which we did not meet in the case of a excitation. Spontaneous decay of atoms in the upper maser level a may be one of the excitation

mechanisms for state b . This could be plausibly represented by replacing the excitation rate density $\lambda_b(z, v, t)$ by $\lambda_b(z, v, t) + f\gamma_a\rho_{aa}(z, v, t)$, where f is the branching ratio (decay from a to b)/(total decay from a) and λ_b now describes only "external" excitation processes.

In order to reduce somewhat the complexity of the subsequent equations we will now proceed as if f were zero. The effects of cascade excitation $a \rightarrow b$ will be discussed by an approximate method in Sec. 20. With this simplification it turns out, as one would expect, that for b excitation $P(b, z, v, t)$ is exactly like (39) except for an over-all sign change and interchange of a and b . Hence, the total polarization $P(z, v, t) = P(a, z, v, t) + P(b, z, v, t)$ is proportional to a quantity

$$N(z, t) = [(\Lambda_a(z, t)/\gamma_a) - (\Lambda_b(z, t)/\gamma_b)], \quad (41)$$

which we will call the "excitation density." This is simply the excess density of active atoms in a steady state in the absence of optical oscillations.

The first-order polarization

$$P^{(1)}(z, t) = \wp \int_{-\infty}^{\infty} dv W(v) [\rho_{ab}^{(1)}(a, z, v, t) \\ + \rho_{ab}^{(1)}(b, z, v, t) + \text{conj.}] \quad (42)$$

is also proportional to $N(z, t)$. For use in Eqs. (13), (14) a spatial Fourier projection on the n th cavity mode is next to be made

$$P_n^{(1)}(t) = (2/L) \int_0^L dz P^{(1)}(z, t) U_n(z). \quad (43)$$

The product $U_n(z)U_\mu(z - v\tau')$ which occurs in (43) may be written as

$$\sin K_n z \sin K_\mu(z - v\tau') \\ = \frac{1}{2} \cos\{(K_n - K_\mu)z + K_\mu v\tau'\} \\ - \frac{1}{2} \cos\{(K_n + K_\mu)z - K_\mu v\tau'\}. \quad (44)$$

The last term will not contribute appreciably to the z integration (43) because the excitation density $N(z, t)$ changes little in an o.f. wavelength. Since the velocity distribution is normally an even function of v only that part of the remainder of (44) which is even in v will contribute to the polarization, i.e.,

$$\frac{1}{2}[\cos(K_n - K_\mu)z] \cos K v \tau',$$

where the subscript μ has been dropped in the last factor since all of the modes considered have very nearly the same wave number $K = \nu/c$.

We find

$$P_n^{(1)}(t) = -\frac{1}{2}i(\wp^2/\hbar) \sum_{\mu} E_\mu \exp -i(\nu_\mu t + \varphi_\mu) N_{n-\mu} \\ \times \int_{-\infty}^{\infty} dv W(v) [\mathfrak{D}(\omega - \nu_\mu + K v)] + \text{conj.}, \quad (45)$$

where

$$\mathfrak{D}(\omega) = 1/(\gamma_{ab} + i\omega) \quad (46)$$

¹⁰ It would be easy to modify the theory to allow the atoms excited to state b to have a different velocity distribution from those excited to state a .

is a convenient abbreviation for a frequently occurring denominator and where

$$N_{n-\mu}(t) = \frac{1}{L} \int_0^L dz N(z, t) \cos \left[(n-\mu) \frac{\pi z}{L} \right] \quad (47)$$

is a spatial Fourier component of the excitation density $N(z, t)$. It should be noted that (45) has a very simple interpretation in terms of Doppler shifts of the atomic transition frequencies by Kv due to the atomic motion. This simplicity will be lost when nonlinear effects are considered.

For the following detailed calculations a Maxwellian distribution

$$W(v) = (u\pi^{1/2})^{-1} \exp(-v^2/u^2) \quad (48)$$

will be assumed. The speed parameter u is related to an effective temperature T by the equation

$$\frac{1}{2}mu^2 = k_B T, \quad (49)$$

where m is the atomic mass and k_B is the Boltzmann constant. If it should develop that a Maxwellian distribution is not realized in practice, some obvious changes in the later work can be made.

With Eq. (48) the integration over v may profitably be done on (39) before that over τ' , and we find

$$P_n^{(1)}(t) = -\frac{1}{2}(\wp^2/\hbar Ku) \times \left[\sum_{\mu=1}^M E_\mu(t) \exp(-i\nu_\mu t + \varphi_\mu(t)) \times N_{n-\mu}(t) Z(\nu_\mu - \omega) + \text{conj.} \right], \quad (50)$$

where $Z(\nu - \omega)$ is an abbreviation for

$$Z(\nu - \omega, \gamma_{ab}, Ku) = iKu \int_0^\infty d\tau \exp\left[i(\nu - \omega)\tau - \gamma_{ab}\tau - \frac{1}{4}K^2u^2\tau^2 \right], \quad (51)$$

which is a complex function well known in the theory of Doppler broadening.¹¹ The function Z is, in fact, a function of a single complex variable ζ

$$Z(\zeta) = 2i \int_{-\infty}^{i\zeta} dt \exp(-t^2 + \zeta^2), \quad (52)$$

where

$$\zeta = \xi + i\eta, \quad (53)$$

with

$$\xi = (\nu - \omega)/Ku, \quad (54)$$

and

$$\eta = \gamma_{ab}/Ku. \quad (55)$$

It is fortunate that extensive tables¹² of the real

¹¹ M. Born, *Optik* (Julius Springer-Verlag, Berlin, 1933), pp. 482-486.

¹² B. D. Fried and S. D. Conte, *The Plasma Dispersion Function (Hilbert Transform of the Gaussian)* (Academic Press, Inc., New York, 1961).

part Z_r and the imaginary part Z_i of $Z(\xi + i\eta)$ are now available.

$P_n^{(1)}(t)$ is a linear function of the complex electric fields $E_\mu(t) \exp(-i\nu_\mu t - i\varphi_\mu(t))$ of the cavity modes and, apart from amplitude modulation arising from a possible slow time variation of the excitation density $N(z, t)$ contains the same frequencies as the cavity field.

To determine amplitude and frequency (or phase) of the oscillations, we write out the contributions of $P_n^{(1)}$ to C_n and S_n of Eqs. (13), (14). These are

$$S_n^{(1)} = -(\wp^2/\hbar Ku) \bar{N} Z_i(\nu_n - \omega) E_n, \quad (56)$$

$$C_n^{(1)} = -(\wp^2/\hbar Ku) \bar{N} Z_r(\nu_n - \omega) E_n, \quad (57)$$

where

$$\bar{N} = N_0(t) = (1/L) \int_0^L dz N(z, t), \quad (58)$$

which will be called the "excitation," is the average of the excitation density over the cavity. We have now reverted to the notation of Eq. (51) for the Z function, but to shorten equations have dropped the parameters γ_{ab} and Ku which appear as arguments in (51).

In this approximation, without nonlinear terms, we can only hope to obtain the condition for starting of oscillations and their frequency at threshold. Furthermore, if the conditions are such that several modes can oscillate, they do so independently of each other and hence can be considered separately. The amplitude equation (14) gives

$$\dot{E}_n = -\frac{1}{2}(\nu/Q_n) E_n - \frac{1}{2}(\nu/\epsilon_0) S_n^{(1)} \quad (59)$$

or for a steady state, for which $\dot{E}_n = 0$,

$$(\wp^2/\epsilon_0 \hbar Ku) \bar{N} Z_i(\nu_n - \omega) = 1/Q_n. \quad (60)$$

To first order in $\eta = \gamma_{ab}/Ku$, we have

$$Z(\xi, \eta) \simeq (1 - 2i\eta\xi) \left[-2 \int_0^\xi e^{x^2} dx + i\pi^{1/2} \right] e^{-\xi^2} - 2i\eta. \quad (61)$$

Hence for pure Doppler line shape the condition (60) for the onset of oscillations in the n th mode may be written as

$$2\pi^{1/2} \left[\frac{e^2}{4\pi\epsilon_0 \hbar u} \right] \times (\wp/e)^2 \lambda \bar{N} \exp(-(\nu_n - \omega)^2/(Ku)^2) = 1/Q_n, \quad (62)$$

where $\lambda = 2\pi/K$ is the wavelength. In these units,

$$e^2/(4\pi\epsilon_0 \hbar c) \approx 1/137 \quad (63)$$

is the fine structure constant, so the left-hand side of (62) is the product of this and four other dimensionless factors: c/u , $2\pi^{1/2}$, $\exp(-(\nu_n - \omega)^2/(Ku)^2)$ and $(\wp/e)^2 \lambda \bar{N}$ which is the net number of active atoms in a cylinder of cross sectional area $(\wp/e)^2$ and length λ . As the frequency detuning $\nu_n - \omega$ increases, the excitation \bar{N} required to initiate oscillations increases in proportion to $\exp+(\nu_n - \omega)^2/(Ku)^2$. The frequency of oscillation is

determined by Eq. (13) in which we may set $\dot{\varphi}_n=0$ without loss of generality. Using Eq. (57), we find

$$\nu_n = \Omega_n + (\nu/2)(\varphi^2 \bar{N} / \epsilon_0 \hbar K u) Z_r(\nu_n - \omega). \quad (64)$$

It is convenient to use the threshold condition (60) to express \bar{N} in terms of Q_n . We find

$$\nu_n - \Omega_n = \frac{1}{2}(\nu/Q_n)[Z_r(\nu_n - \omega)/Z_i(\nu_n - \omega)] \quad (65)$$

or in the approximation $\gamma_{ab}/Ku \ll 1$

$$\nu_n - \Omega_n = -\left(\frac{1}{\pi^{1/2}}\right)\left(\frac{\nu}{Q_n}\right) \int_0^{\xi_n} dx e^{x^2}. \quad (66)$$

If the integral (66) is expanded to first order in $\xi_n = (\nu_n - \omega)/Ku$ we obtain

$$(\nu_n - \Omega_n)/(\omega - \nu_n) \approx (1/\pi^{1/2})(\nu/QKu) = S, \quad (67)$$

which implies "linear pulling," i.e., the detuning of ν_n from the cavity frequency Ω_n is proportional to the amount $\omega - \nu_n$ by which the oscillator frequency ν_n is removed from the atomic resonance frequency ω . The right-hand side of (67) (S = "stabilization factor"¹³) is about 1/800 for typical values of the parameters used in Sec. 2.

The more accurate expression (65) indicates a "non-linear pulling" such that the oscillator frequency is

nearer to the atomic frequency than it would be for linear pulling. To the next order in $(\Omega_n - \omega)/Ku$, but still for $\gamma_{ab} \ll Ku$, we find

$$(\nu_n - \Omega_n)/(\omega - \nu_n) \approx [1 + \frac{1}{3}(\Omega_n - \omega)^2/(Ku)^2]S. \quad (68)$$

7. THIRD-ORDER TERMS

We now carry out a similar calculation for the third-order quantity $P_n^{(3)}(t)$ using Eqs. (8), (23), (30), (31) and (32). The integration of $\lambda_a(z, v, t_0) \rho_{ab}^{(3)}(a, z, t_0, v, t)$ over times t_0 of excitation involves integrals like

$$\int_{-\infty}^t dt_0 \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt''' F(t, t', t'', t''') \exp -\gamma_a(t''' - t_0),$$

which by a repeated interchange of orders of integration can be reduced to

$$(1/\gamma_a) \int_{-\infty}^t dt' \int_{-\infty}^{t'} dt'' \int_{-\infty}^{t''} dt''' F(t, t', t'', t''').$$

Again, we keep only exponential factors in the time integration which are able to have resonance, and find after changes of variables

$$\tau' = t - t', \quad \tau'' = t' - t'', \quad \tau''' = t'' - t''' \quad (69)$$

and some algebraic manipulations

$$\begin{aligned} \rho_{ab}^{(3)}(z, v, t) = & \frac{1}{8} i \varphi^3 \hbar^{-3} N(z, t) \sum_{\mu} \sum_{\rho} \sum_{\sigma} E_{\mu} E_{\rho} E_{\sigma} \left\{ [\exp -i(\nu_{\mu} t + \varphi_{\mu}) + i(\nu_{\rho} t + \varphi_{\rho}) - i(\nu_{\sigma} t + \varphi_{\sigma})] \right. \\ & \times \int_0^{\infty} d\tau' \int_0^{\infty} d\tau'' \int_0^{\infty} d\tau''' U_{\mu}(z - v\tau') U_{\rho}(z - v\tau' - v\tau'') U_{\sigma}(z - v\tau' - v\tau'' - v\tau''') \\ & \times \{ \exp - [(\gamma_{ab} - i\nu_{\mu} + i\nu_{\rho} - i\nu_{\sigma} + i\omega)\tau' + (\gamma_a + i\nu_{\rho} - i\nu_{\sigma})\tau'' + (\gamma_{ab} + i\omega - i\nu_{\sigma})\tau'''] \} \\ & + [\exp -i(\nu_{\mu} t + \varphi_{\mu}) - i(\nu_{\rho} t + \varphi_{\rho}) + i(\nu_{\sigma} t + \varphi_{\sigma})] \\ & \times \int_0^{\infty} d\tau' \int_0^{\infty} d\tau'' \int_0^{\infty} d\tau''' U_{\mu}(z - v\tau') U_{\rho}(z - v\tau' - v\tau'') U_{\sigma}(z - v\tau' - v\tau'' - v\tau''') \\ & \times [\exp - [(\gamma_{ab} - i\nu_{\mu} - i\nu_{\rho} + i\nu_{\sigma} + i\omega)\tau' + (\gamma_a - i\nu_{\rho} + i\nu_{\sigma})\tau'' + (\gamma_{ab} - i\omega + i\nu_{\sigma})\tau''']] \} \\ & + \text{same with } a \text{ and } b \text{ interchanged.} \quad (70) \end{aligned}$$

In calculation of the Fourier projection $P_n^{(3)}(t)$ integrals of the form

$$\left(\frac{2}{L}\right) \int_0^L dz N(z) U_n(z) U_{\mu}(z - v\tau') U_{\rho}(z - v\tau' - v\tau'') U_{\sigma}(z - v\tau' - v\tau'' - v\tau''')$$

appear. The product of the four sine functions can be reduced to

$$\begin{aligned} \frac{1}{8} [& \cos(K_{\rho} - K_{\sigma} + K_{\mu} - K_n)z \cos Kv(\tau''' - \tau') + \cos(K_{\rho} - K_{\sigma} - K_{\mu} + K_n)z \cos Kv(\tau''' + \tau') \\ & + \cos(K_{\rho} + K_{\sigma} - K_{\mu} - K_n)z \cos Kv(\tau' + 2\tau'' + \tau''')] \end{aligned}$$

¹³ The term "stabilization factor" (\approx cavity band width/atom bandwidth) was previously used in a theory of the ammonia beam maser (Ref. 8, p. 460) where its numerical value was large compared to unity.

apart from rapidly oscillating terms and those odd in v . The integration over v may now be carried out, and we find

$$\begin{aligned}
 P_n^{(3)}(t) = & \frac{1}{3^2} i \varphi^4 \hbar^{-3} \sum_{\mu} \sum_{\rho} \sum_{\sigma} E_{\mu} E_{\rho} E_{\sigma} \left\{ \left[\exp -i(\nu_{\mu} t + \varphi_{\mu}) + i(\nu_{\rho} t + \varphi_{\rho}) - i(\nu_{\sigma} t + \varphi_{\sigma}) \right] \right. \\
 & \times \int_0^{\infty} d\tau' \int_0^{\infty} d\tau'' \int_0^{\infty} d\tau''' \left[N_{(\rho-\sigma+\mu-n)} \exp -\frac{1}{4}(Ku)^2(\tau''' - \tau')^2 + N_{(\rho-\sigma-\mu+n)} \exp -\frac{1}{4}(Ku)^2(\tau''' + \tau')^2 \right. \\
 & + N_{(\rho+\sigma-\mu-n)} \exp -\frac{1}{4}(Ku)^2(\tau' + 2\tau'' + \tau''')^2 \left. \right] \left[\exp -(\gamma_{ab} - i\nu_{\mu} + i\nu_{\rho} - i\nu_{\sigma} + i\omega)\tau' \right. \\
 & - (\gamma_a + i\nu_{\rho} - i\nu_{\sigma})\tau'' - (\gamma_{ab} + i\omega - i\nu_{\sigma})\tau'''] + \left. \left[\exp -i(\nu_{\mu} t + \varphi_{\mu}) - i(\nu_{\rho} t + \varphi_{\rho}) + i(\nu_{\sigma} t + \varphi_{\sigma}) \right] \right. \\
 & \times \int_0^{\infty} d\tau' \int_0^{\infty} d\tau'' \int_0^{\infty} d\tau''' \left[N_{(\rho-\sigma+\mu-n)} \exp -\frac{1}{4}(Ku)^2(\tau''' - \tau')^2 + N_{(\rho-\sigma-\mu+n)} \exp -\frac{1}{4}(Ku)^2(\tau''' + \tau')^2 \right. \\
 & + N_{(\rho+\sigma-\mu-n)} \exp -\frac{1}{4}(Ku)^2(\tau' + 2\tau'' + \tau''')^2 \left. \right] \left[\exp -(\gamma_{ab} - i\nu_{\mu} - i\nu_{\rho} + i\nu_{\sigma} + i\omega)\tau' - (\gamma_a - i\nu_{\rho} + i\nu_{\sigma})\tau'' \right. \\
 & \left. \left. - (\gamma_{ab} - i\omega + i\nu_{\sigma})\tau'''] \right\} + \text{same with } \gamma_a \text{ and } \gamma_b \text{ interchanged} + \text{complex conjugate.} \quad (71)
 \end{aligned}$$

One of the most important characteristics of the third-order polarization is that it has constituents which oscillate at all possible frequencies $\nu_{\mu} \mp \nu_{\rho} \pm \nu_{\sigma}$. The combination of signs appearing here is correlated with use of the rotating wave approximation. Terms with frequencies such as $\nu_{\mu} + \nu_{\rho} + \nu_{\sigma}$ (near the third harmonic) are thereby neglected.

The formidable expression (71) can be simplified in either of two limiting cases: (a) no atomic motion ($u=0$), or (b) "Doppler limit," i.e., Ku much larger than γ_{ab} and various frequency differences such as $\nu_{\mu} + \nu_{\rho} - 2\nu_{\sigma}$, etc. In the absence of atomic motion, one finds after some rewriting

$$\begin{aligned}
 P_n^{(3)}(t) = & \frac{1}{3^2} i \hbar^{-3} \varphi^4 \sum_{\mu} \sum_{\rho} \sum_{\sigma} E_{\mu} E_{\rho} E_{\sigma} \left[\exp -i(\nu_{\mu} t + \varphi_{\mu}) + i(\nu_{\rho} t + \varphi_{\rho}) - i(\nu_{\sigma} t + \varphi_{\sigma}) \right] \\
 & \times \left[N_{(\rho-\sigma+\mu-n)} + N_{(\rho-\sigma-\mu+n)} + N_{(\rho+\sigma-\mu-n)} \right] \cdot \mathfrak{D}(\nu_{\rho} - \nu_{\mu} - \nu_{\sigma} + \omega) \\
 & \times \left[\mathfrak{D}_a(\nu_{\rho} - \nu_{\sigma}) + \mathfrak{D}_b(\nu_{\rho} - \nu_{\sigma}) \right] \left[\mathfrak{D}(\omega - \nu_{\sigma}) + \mathfrak{D}(\nu_{\rho} - \omega) \right] + \text{conjugate,} \quad (72)
 \end{aligned}$$

where $\mathfrak{D}(\omega)$ was defined by Eq. (46) and

$$\mathfrak{D}_\alpha(\omega) = 1/(\gamma_\alpha + i\omega), \quad \alpha = a, b. \quad (73)$$

The "Doppler limit" is appropriate for many possible gaseous optical masers, and for most of the remainder of this paper we will be dealing only with this case. Then

$$\exp -\frac{1}{4}(Ku)^2(\tau''' - \tau')^2$$

acts like a delta function of $\tau''' - \tau'$ and the integration over τ''' can be done in the form

$$\int_0^{\infty} d\tau' \int_0^{\infty} d\tau''' G(\tau', \tau''') \exp -\frac{1}{4}(Ku)^2(\tau''' - \tau')^2 \simeq \frac{2\pi^{1/2}}{Ku} \int_0^{\infty} d\tau' G(\tau', \tau'). \quad (74)$$

The other Gaussian factors do not have their full peaks in the range of integration, and give contributions which we neglect because they lead to expressions with higher powers of Ku in the denominator. Then, after performing the simple integrations over τ' and τ'' , we find

$$\begin{aligned}
 P_n^{(3)}(t) = & \frac{1}{3^2} i \pi^{1/2} \left[\varphi^4 / (\hbar^3 Ku) \right] \sum_{\mu} \sum_{\rho} \sum_{\sigma} E_{\mu} E_{\rho} E_{\sigma} N_{(\rho-\sigma+\mu-n)} \left\{ \mathfrak{D}(\omega - \frac{1}{2}\nu_{\mu} + \frac{1}{2}\nu_{\rho} - \nu_{\sigma}) \left[\mathfrak{D}_a(\nu_{\rho} - \nu_{\sigma}) + \mathfrak{D}_b(\nu_{\rho} - \nu_{\sigma}) \right] \right. \\
 & \times \left[\exp -i(\nu_{\mu} - \nu_{\rho} + \nu_{\sigma})t - i(\varphi_{\mu} - \varphi_{\rho} + \varphi_{\sigma}) \right] + \mathfrak{D}(-\frac{1}{2}\nu_{\mu} - \frac{1}{2}\nu_{\rho} + \nu_{\sigma}) \left[\mathfrak{D}_a(\nu_{\sigma} - \nu_{\rho}) + \mathfrak{D}_b(\nu_{\sigma} - \nu_{\rho}) \right] \\
 & \left. \times \left[\exp[-i(\nu_{\mu} + \nu_{\rho} - \nu_{\sigma})t - i(\varphi_{\mu} + \varphi_{\rho} - \varphi_{\sigma})] \right] \right\} + \text{complex conjugate.} \quad (75)
 \end{aligned}$$

By interchanging ρ and σ in the second group of terms, this may be written more compactly as

$$\begin{aligned}
 P_n^{(3)}(t) = & \frac{1}{3^2} i \pi^{1/2} \left[\varphi^4 / (\hbar^3 Ku) \right] \sum_{\mu} \sum_{\rho} \sum_{\sigma} E_{\mu} E_{\rho} E_{\sigma} \left[\exp[-i(\nu_{\mu} - \nu_{\rho} + \nu_{\sigma})t - i(\varphi_{\mu} - \varphi_{\rho} + \varphi_{\sigma})] \right] \\
 & \times \left[N_{(\rho-\sigma+\mu-n)} \mathfrak{D}(\omega - \frac{1}{2}\nu_{\mu} + \frac{1}{2}\nu_{\rho} - \nu_{\sigma}) + N_{(\sigma-\rho+\mu-n)} \mathfrak{D}(-\frac{1}{2}\nu_{\mu} - \frac{1}{2}\nu_{\sigma} + \nu_{\rho}) \right] \left[\mathfrak{D}_a(\nu_{\rho} - \nu_{\sigma}) + \mathfrak{D}_b(\nu_{\rho} - \nu_{\sigma}) \right] \\
 & + \text{complex conjugate.} \quad (76)
 \end{aligned}$$

8. SINGLE FREQUENCY OPERATION

For this case, the triple summation over μ , ρ , and σ reduces to a single term

$$P_n^{(3)}(t) = \frac{1}{16} i \pi^{1/2} \bar{N} [\varphi^4 / (\hbar^3 \gamma_a \gamma_b K u)] \\ \times \gamma_{ab} [\mathfrak{D}(\omega - \nu_n) + \mathfrak{D}(0)] E_n^{(3)} \exp -i(\nu_n t + \varphi_n) \\ + \text{complex conjugate.} \quad (77)$$

For use in Eqs. (13), (14) we need the in-phase and quadrature components of this

$$C_n^{(3)}(t) = \frac{1}{8} \pi^{1/2} \bar{N} [\varphi^4 / (\hbar^3 \gamma_a \gamma_b K u)] \\ \times \gamma_{ab} (\omega - \nu_n) \mathfrak{L}(\omega - \nu_n) E_n^3 \quad (78)$$

and

$$S_n^{(3)}(t) = \frac{1}{8} \pi^{1/2} \bar{N} [\varphi^4 / (\hbar^3 \gamma_a \gamma_b K u)] \\ \times [1 + \gamma_{ab}^2 \mathfrak{L}(\omega - \nu_n)] E_n^3, \quad (79)$$

where the Lorentzian function is denoted by

$$\mathfrak{L}(\omega - \nu) = [\gamma_{ab}^2 + (\omega - \nu)^2]^{-1}. \quad (80)$$

The amplitude determining Eq. (13) is then of the form¹⁴

$$\dot{E}_n = \alpha_n E_n - \beta_n E_n^3, \quad (81)$$

where

$$\alpha_n = -\frac{1}{2} (\nu / Q_n) + \frac{1}{2} \nu \bar{N} [\varphi^2 / (\epsilon_0 \hbar K u)] Z_i(\nu_n - \omega), \quad (82)$$

$$\beta_n = (\nu / 16) \pi^{1/2} \bar{N} [\varphi^4 / (\epsilon_0 \hbar^3 \gamma_a \gamma_b K u)] \\ \times [1 + \gamma_{ab}^2 \mathfrak{L}(\nu_n - \omega)]. \quad (83)$$

It is useful to employ the starting condition (60) to express the coefficients α_n , β_n (and others which appear later) in terms of a ratio

$$\mathfrak{X} = \bar{N} / \bar{N}_T \quad (84)$$

called the "relative excitation" where \bar{N}_T is the excitation required for threshold oscillations when the cavity frequency Ω_n is tuned to the peak ω of the atomic resonance curve. We find

$$\alpha_n = \frac{1}{2} (\nu / Q_n) \{ [Z_i(\nu_n - \omega) / Z_i(0)] \mathfrak{X} - 1 \} \quad (85)$$

and

$$\beta_n = \frac{1}{16} \pi^{1/2} (\nu / Q_n) [\mathfrak{X} \varphi^2 / (\hbar^3 \gamma_a \gamma_b Z_i(0))] \\ \times [1 + \gamma_{ab}^2 \mathfrak{L}(\nu_n - \omega)]. \quad (86)$$

A stable steady state occurs for an intensity of oscillations

$$E_n^2 = \alpha_n / \beta_n, \quad (87)$$

which is easily related to the relative excitation with the help of Eqs. (85) and (86).

The frequency determining equation (again $\dot{\varphi}_n$ may

¹⁴ It is easy to see that the coefficient α_n is simply related to the gain (negative absorption) coefficient of the medium at frequency ν_n for small signals. However, the gain coefficient for a strong signal cannot be safely inferred from Eq. (81), since standing rather than traveling waves were assumed in its derivation. A theory of a traveling wave maser along the lines of this paper will be given later.

be taken zero) is

$$\nu_n = \Omega_n - \frac{1}{2} (\nu / (\epsilon_0 E_n)) [C_n^{(1)} + C_n^{(3)}]. \quad (88)$$

Since the frequency of oscillation ν_n will differ little from the cavity resonance frequency Ω_n , the right side of Eq. (88) may, to a sufficiently good approximation, be evaluated for $\nu_n = \Omega_n$. We may then write

$$\nu_n = \Omega_n + \sigma_n + \rho_n E_n^2, \quad (89)$$

where

$$\sigma_n = \frac{1}{2} \nu [\varphi^2 \bar{N} / (\epsilon_0 \hbar K u)] Z_r(\Omega_n - \omega) \quad (90)$$

and

$$\rho_n = +\frac{1}{16} \pi^{1/2} \nu [\varphi^4 \bar{N} / (\epsilon_0 \hbar^3 \gamma_a \gamma_b K u)] \\ \times \gamma_{ab} (\Omega_n - \omega) \mathfrak{L}(\Omega_n - \omega) \quad (91)$$

or expressing these coefficients in terms of the relative excitation (84)

$$\sigma_n = \frac{1}{2} (\nu / Q_n) \mathfrak{X} Z_r(\Omega_n - \omega) / Z_i(0) \quad (92)$$

and

$$\rho_n = +\frac{1}{16} \pi^{1/2} (\nu / Q_n) \mathfrak{X} [\varphi^2 / (\hbar^3 \gamma_a \gamma_b Z_i(0))] \\ \times \gamma_{ab} (\Omega_n - \omega) \mathfrak{L}(\Omega_n - \omega) \quad (93)$$

so that

$$\rho_n E_n^2 = \rho_n \alpha_n / \beta_n \\ = \frac{1}{2} (\nu / Q_n) \{ [Z_i(\Omega_n - \omega) / Z_i(0)] \mathfrak{X} - 1 \} \gamma_{ab} (\Omega_n - \omega) / \\ [2\gamma_{ab}^2 + (\Omega_n - \omega)^2]. \quad (94)$$

The frequency now depends on the relative excitation (and hence on the power level) as well as on the detuning, i.e., there is frequency "pushing" as well as "pulling."

Equations (89), (92), and (94) indicate that for

$$(\Omega_n - \omega)^2 + 2\gamma_{ab}^2 < -\gamma_{ab} (\Omega_n - \omega) Z_i(\Omega_n - \omega) / \\ Z_r(\Omega_n - \omega) \quad (95)$$

an increase of excitation should move the frequency in the direction from ω toward Ω_n . For small detuning the right side of (95) is approximately $\frac{1}{2} \pi^{1/2} \gamma_{ab} K u$.

The dependence of power level on excitation and detuning is given by Eq. (87). Using the value of $Z_i(\xi_n, \eta)$ for $\mathfrak{X} = 0$, this equation may be written approximately as

$$(\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b) \\ = 8 \left\{ \left[\exp -\frac{(\Omega_n - \omega)^2}{(K u)^2} \right] - \mathfrak{X}^{-1} \right\} / [1 + \gamma_{ab}^2 \mathfrak{L}(\Omega_n - \omega)]. \quad (96)$$

This expression agrees with the linear approximation (62) in predicting threshold for relative excitation

$$\mathfrak{X} = \bar{N} / \bar{N}_T = \exp(\Omega_n - \omega)^2 / (K u)^2 \quad (97)$$

when there is detuning. Because of its derivation from a third order perturbation theory, Eq. (96) should not be trusted unless it predicts a value of the "saturation

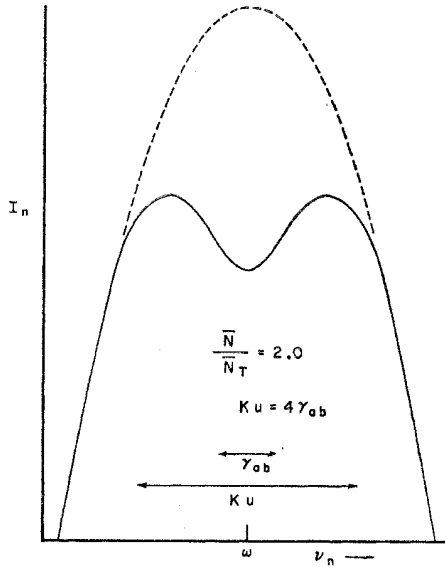


FIG. 2. Relative intensity of oscillation as a function of detuning. The solid curve, drawn for parameters $\bar{N} = 2\bar{N}_T$ and $Ku = 4\gamma_{ab}$ represents Eq. (96). The dotted curve indicates the Doppler gain profile of the numerator of (96).

parameter¹⁵

$$I_n = \frac{1}{2} (\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b) \quad (98)$$

much less than unity.

The numerator in Eq. (96) has a peak for resonant tuning $\Omega_n = \omega$, but the denominator, which comes from the nonlinear term involving the coefficient β_n also has a peak when $\Omega_n = \omega$. Under certain conditions, the over-all curve of $E_n^2 = \alpha_n / \beta_n$ versus detuning $\Omega_n - \omega$ should have a flattened peak at resonance, or even a dip between two maxima. The condition for the appearance of two maxima is

$$(\gamma_{ab}/Ku)^2 < \frac{1}{2} \{1 - \exp[-(\Omega_n^* - \omega)^2 / (Ku)^2]\}, \quad (99)$$

where $\Omega_n^* - \omega$ is the detuning required to stop oscillations at the given level of excitation. The double peak¹⁶ (see Fig. 2) should thus be seen somewhat above threshold, i.e., for relative excitation

$$\mathfrak{R} > 1 / [1 - 2(\gamma_{ab}/(Ku))^2]. \quad (100)$$

Under this degree of relative excitation, the electric field at central tuning is given by

$$I_n = \frac{1}{2} (\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b) \approx 4(\gamma_{ab}/(Ku))^2 \ll 1 \quad (101)$$

so that the neglect of higher orders of perturbation theory should not be too serious, provided, as we assume, that $\gamma_{ab} \ll Ku$.

¹⁵ The significance of this quantity is shown more clearly in Sec. 18.

¹⁶ This dip has recently been observed. R. A. McFarlane, W. R. Bennett, and W. E. Lamb, *Appl. Phys. Letters* **2**, 189 (1963); A. Szöke and A. Javan, *Phys. Rev. Letters* **10**, 521 (1963).

TABLE I. This shows the twenty-seven possible values of the summation indices μ, ρ, σ which appear in Eq. (76) for "three"-mode oscillation. The fourth column gives the corresponding frequencies $\nu_\mu - \nu_\rho + \nu_\sigma$. The last column contains numerical values for the amount by which these frequencies exceed ν_1 . We have taken $\nu_2 - \nu_1 = 150$ Mc/sec and $\nu_3 - \nu_2 = 151$ Mc/sec in order to simulate (but greatly to exaggerate) nonlinear pulling effects.

μ	ρ	σ	$\nu_\mu - \nu_\rho + \nu_\sigma$	Typical $\delta\nu$ (Mc/sec)
1	1	1	ν_1	0
1	1	2	ν_2	150
1	1	3	ν_3	301
1	2	1	$2\nu_1 - \nu_2$	-150
1	2	2	ν_1	0
1	2	3	$\nu_1 + \nu_3 - \nu_2$	151
1	3	1	$2\nu_1 - \nu_3$	-301
1	3	2	$\nu_1 + \nu_2 - \nu_3$	-151
1	3	3	ν_1	0
2	1	1	ν_2	150
2	1	2	$2\nu_2 - \nu_1$	300
2	1	3	$\nu_2 + \nu_3 - \nu_1$	451
2	2	1	ν_1	0
2	2	2	ν_2	150
2	2	3	ν_3	301
2	3	1	$\nu_1 + \nu_2 - \nu_3$	-151
2	3	2	$2\nu_2 - \nu_3$	-1
2	3	3	ν_2	150
3	1	1	ν_3	301
3	1	2	$\nu_2 + \nu_3 - \nu_1$	451
3	1	3	$2\nu_3 - \nu_1$	602
3	2	1	$\nu_1 + \nu_3 - \nu_2$	151
3	2	2	ν_3	301
3	2	3	$2\nu_3 - \nu_2$	452
3	3	1	ν_1	0
3	3	2	ν_2	150
3	3	3	ν_3	301

The physical interpretation of the dip is discussed in Secs. 17 and 18 from several points of view.

9. MULTIPLE MODE OPERATION

As the excitation is increased beyond that required for threshold of single frequency oscillation, other frequencies appear in the output of an optical maser oscillator. We wish to use the expression (76) for the nonlinear polarization in the electromagnetic field equations (13), (14) in order to account for the observed phenomena.

The first theory of an oscillator capable of multi-frequency operation was given by van der Pol¹⁷ in 1921-22. The necessary nonlinear features were provided by cubic terms in the current-voltage characteristic of a triode vacuum tube. The tank had two $R-L-C$ circuits with resonance frequencies Ω_1 and Ω_2 . Van der Pol found that steady oscillations could occur only at frequencies near Ω_1 or Ω_2 , but that simultaneous steady operation at the two frequencies was impossible. There were hysteresis phenomena, i.e., the choice of steady state of oscillation depended on the past history of the circuit parameters.

Multicavity magnetrons provide very important

¹⁷ B. van der Pol, *Phil. Mag.* **43**, 700 (1922) and a review article *Proc. Inst. Radio Engrs.* **22**, 1051 (1934).

TABLE II. This table gives various quantities needed for the evaluation of Eq. (76) for three-frequency oscillation as described in the text.

μ	ρ	σ	n	$\nu_\rho - \nu_\sigma$	$\omega - \frac{1}{2}\nu_\mu + \frac{1}{2}\nu_\rho - \nu_\sigma$	$-\frac{1}{2}\nu_\mu - \frac{1}{2}\nu_\sigma + \nu_\rho$	$\nu_\mu - \nu_\rho + \nu_\sigma$	$\rho - \sigma + \mu - n$	$\sigma - \rho + \mu - n$
1	1	1	1	0	$\omega - \nu_1$	0	ν_1	0	0
1	2	2	1	0	$\omega - \nu_{12}$	$\frac{1}{2}\Delta$	ν_1	0	0
1	3	3	1	0	$\omega - \nu_2$	Δ	ν_1	0	0
2	2	1	1	Δ	$\omega - \nu_1$	$\frac{1}{2}\Delta$	ν_1	2	0
3	3	1	1	2Δ	$\omega - \nu_1$	Δ	ν_1	4	0
2	3	2	1	Δ	$\omega - \nu_{12}$	Δ	$2\nu_2 - \nu_3$	2	0
2	2	2	2	0	$\omega - \nu_2$	0	ν_2	0	0
1	1	2	2	$-\Delta$	$\omega - \nu_2$	$-\frac{1}{2}\Delta$	ν_2	-2	0
2	1	1	2	0	$\omega - \nu_{12}$	$-\frac{1}{2}\Delta$	ν_2	0	0
2	3	3	2	0	$\omega - \nu_{23}$	$\frac{1}{2}\Delta$	ν_2	0	0
3	3	2	2	Δ	$\omega - \nu_2$	$\frac{1}{2}\Delta$	ν_2	2	0
1	2	3	2	$-\Delta$	$\omega - \nu_{23}$	"0"	$\nu_1 + \nu_3 - \nu_2$	-2	0
3	2	1	2	Δ	$\omega - \nu_{12}$	"0"	$\nu_1 + \nu_3 - \nu_2$	2	0
3	3	3	3	0	$\omega - \nu_3$	0	ν_3	0	0
1	1	3	3	-2Δ	$\omega - \nu_3$	$-\Delta$	ν_3	-4	0
2	2	3	3	$-\Delta$	$\omega - \nu_3$	$-\frac{1}{2}\Delta$	ν_3	-2	0
3	1	1	3	0	$\omega - \nu_2$	$-\Delta$	ν_3	0	0
3	2	2	3	0	$\omega - \nu_{23}$	$-\frac{1}{2}\Delta$	ν_3	0	0
2	1	2	3	$-\Delta$	$\omega - \nu_{23}$	$-\Delta$	$2\nu_2 - \nu_1$	-2	0

examples of oscillators capable of multifrequency operation, and here again the normal pattern is that an oscillation existing at one frequency tends to suppress one at another frequency.

There is, of course, a very close connection between van der Pol's work and ours. Where he dealt with a nonlinear triode characteristic for a vacuum tube, we are concerned with the nonlinear response of an assembly of atomic systems which obey the laws of quantum mechanics. A one-to-one correspondence can be set up between the two problems. The Fabry-Perot cavity modes correspond to the resonant constituents of van der Pol's plate circuit. As we will see, however, the effective tube characteristics of the optical medium differ qualitatively from that assumed by van der Pol, and hence the optical maser behaves in a very different fashion with respect to multifrequency operation than the oscillator in van der Pol's original model.

Rather than deal next with the case of two-frequency operation, it will perhaps save space to consider first the more general case of three-frequency operation. Probably most of the interesting phenomena for optical gaseous masers can be understood without dealing explicitly with more than three frequencies. After the more general equations have been obtained, we can easily drop terms and discuss two frequency oscillation as a special and simpler case.

We consider the expression (76) for the third-order polarization, in which the indices μ, ρ, σ , and n can each take on values 1, 2, and 3. It is useful to have the ingredients of the summands in tabular form. The entries in the fourth column of Table I give the frequencies of the various summands in $P_n^{(3)}$ identified by the μ, ρ , and σ values in the first three columns. It will be noted that besides the three frequencies ν_1, ν_2 , and ν_3 assumed in the cavity excitation, there are nine

additional frequencies present in the polarization of the medium. Hence there must be fields in the cavity at these new frequencies, and the desired self-consistency for three-frequency operation is in jeopardy. However, under certain conditions which will be determined later, the fields at the new frequencies do not produce appreciable effects, even though the frequencies lie close to cavity resonance, so that the calculation can be made as planned.

As is already apparent from the single-frequency case, the oscillation frequencies ν_n are typically very close to the cavity frequencies Ω_n , which are equally spaced and separated by

$$\Delta \approx 150 \text{ Mc/sec.} \tag{102}$$

Hence three of the new frequencies: $2\nu_2 - \nu_3, \nu_1 + \nu_3 - \nu_2$, and $2\nu_2 - \nu_1$ are very close to the three main frequencies ν_1, ν_2 , and ν_3 , respectively, and the corresponding terms are carried along in the calculations. The remaining frequencies can be ignored as long as the oscillator is appreciably below threshold for four-frequency operation.

Any pulling of ν_n from Ω_n is typically measured in kc/sec and hence the ν_n are not detuned from Ω_n by an appreciable fraction either of the cavity bandwidths ν/Q_n which are about 1 Mc/sec, or of the radiative decay constants γ_a, γ_b , or γ_{ab} which may be 10 Mc/sec or more. The entries in columns 5, 6, and 7 of Table II occur in the frequency denominators in Eq. (76) and are there to be combined with imaginary numbers $-i\gamma_a, -i\gamma_b$, or $-i\gamma_{ab}$. We have neglected the small terms arising from frequency pulling. Thus $\nu_2 - \nu_1$ is freely replaced by Δ of Eq. (102), etc. A symbol like ν_{12} denotes a frequency halfway between ν_1 and ν_2 , etc.,

$$\nu_{12} = \frac{1}{2}(\nu_1 + \nu_2), \text{ etc.} \tag{103}$$

The entries in the last two columns of Table II are the

integers characterizing the spatial Fourier components (47) of the excitation density which are needed for the evaluation of the contribution to (76) arising from the indices μ, ρ, σ , and n .

The combinations (μ, ρ, σ) which contribute third order polarizations at frequency ν_1 are (1,1,1), (1,2,2), (1,3,3), (2,2,1), and (3,3,1) while (2,3,2) gives a con-

tribution near ν_1 . Each summand has a product of two-resonance denominators. The dominant terms are (1,1,1), (1,2,2), and (1,3,3) since they do not necessarily contain inverse powers of Δ . The other terms have at least one power of Δ in the denominator. All terms will ultimately be expanded to order $1/\Delta^2$.

We find

$$P_1^{(3)}(t) = \frac{1}{16} i \pi^{1/2} [\varphi^4 / \hbar^3 K u] \{ [E_1^3 \bar{N}(\gamma_{ab} / (\gamma_a \gamma_b)) (\mathfrak{D}(\omega - \nu_1) + \mathfrak{D}(0)) + E_1 E_2^2 \bar{N}(\gamma_{ab} / (\gamma_a \gamma_b)) (\mathfrak{D}(\omega - \nu_{12}) + \mathfrak{D}(\Delta/2)) + E_1 E_3^2 \bar{N}(\gamma_{ab} / (\gamma_a \gamma_b)) (\mathfrak{D}(\omega - \nu_2) + \mathfrak{D}(\Delta)) + E_2^2 E_1 [\frac{1}{2} N_2 \mathfrak{D}(\omega - \nu_1) + \frac{1}{2} \bar{N} \mathfrak{D}(\Delta/2)] [\mathfrak{D}_a(\Delta) + \mathfrak{D}_b(\Delta)] + E_3^2 E_1 [\frac{1}{2} N_4 \mathfrak{D}(\omega - \nu_1) + \frac{1}{2} \bar{N} \mathfrak{D}(\Delta)] \times [\mathfrak{D}_a(2\Delta) + \mathfrak{D}_b(2\Delta)] \} \exp -i(\nu_1 t + \varphi_1) + \frac{1}{2} E_2^2 E_3 [N_2 \mathfrak{D}(\omega - \nu_{12}) + \bar{N} \mathfrak{D}(\Delta)] [\mathfrak{D}_a(\Delta) + \mathfrak{D}_b(\Delta)] \times \exp -i((2\nu_2 - \nu_3)t - (2\varphi_2 - \varphi_3)) \} + \text{complex conjugate}. \quad (104)$$

The in-phase coefficient $C_1^{(3)}$ is then

$$C_1^{(3)} = \frac{1}{8} \pi^{1/2} [\varphi^4 / (\hbar^3 K u)] \{ [E_1^3 \bar{N}(\gamma_{ab} / (\gamma_a \gamma_b)) (\omega - \nu_1) \mathfrak{L}(\omega - \nu_1) + E_1 E_2^2 \bar{N}(\gamma_{ab} / (\gamma_a \gamma_b)) [(\omega - \nu_{12}) \mathfrak{L}(\omega - \nu_{12}) + (2/\Delta)] + E_1 E_3^2 \bar{N}(\gamma_{ab} / (\gamma_a \gamma_b)) [(\omega - \nu_2) \mathfrak{L}(\omega - \nu_2) + (1/\Delta)] + E_2^2 E_1 N_2 \gamma_{ab} (\omega - \nu_1 + \Delta) \mathfrak{L}(\omega - \nu_1) / \Delta^2 + \frac{1}{4} E_3^2 E_1 N_4 \gamma_{ab} (\omega - \nu_1 + 2\Delta) \mathfrak{L}(\omega - \nu_1) / \Delta^2 + E_2^2 E_3 \Delta^{-2} \{ N_2 \gamma_{ab} (\omega - \nu_{12} + \Delta) \mathfrak{L}(\omega - \nu_{12}) \cos \psi + [N_2 (\gamma_{ab}^2 - (\omega - \nu_{12}) \Delta) \mathfrak{L}(\omega - \nu_{12}) - \bar{N}] \sin \psi \} \}, \quad (105)$$

where the "relative phase angle" ψ is defined as

$$\psi = (2\nu_2 - \nu_1 - \nu_3)t + (2\varphi_2 - \varphi_1 - \varphi_3). \quad (106)$$

The quadrature coefficient $S_1^{(3)}$ is given by

$$S_1^{(3)} = \frac{1}{8} \pi^{1/2} [\varphi^4 / (\hbar^3 K u)] \{ [E_1^3 \bar{N}[\gamma_{ab}^2 \mathfrak{L}(\omega - \nu_1) + 1] / (\gamma_a \gamma_b) + E_1 E_2^2 \bar{N}[\gamma_{ab}^2 \mathfrak{L}(\omega - \nu_{12}) + 4\gamma_{ab}^2 \Delta^{-2}] / (\gamma_a \gamma_b) + E_1 E_3^2 \bar{N}[\gamma_{ab}^2 \mathfrak{L}(\omega - \nu_2) + \gamma_{ab}^2 \Delta^{-2}] / (\gamma_a \gamma_b) + E_2^2 E_1 \Delta^{-2} [N_2 (\gamma_{ab}^2 - (\omega - \nu_1) \Delta) \mathfrak{L}(\omega - \nu_1) - 2\bar{N}] + \frac{1}{4} E_3^2 E_1 \Delta^{-2} [N_4 (\gamma_{ab}^2 - 2(\omega - \nu_1) \Delta) \mathfrak{L}(\omega - \nu_1) - 2\bar{N}] + E_2^2 E_3 \Delta^{-2} \{ [N_2 (\gamma_{ab}^2 - (\omega - \nu_{12}) \Delta) \mathfrak{L}(\omega - \nu_{12}) - \bar{N}] \cos \psi - N_2 \gamma_{ab} (\omega - \nu_{12} + \Delta) \mathfrak{L}(\omega - \nu_{12}) \sin \psi \} \}. \quad (107)$$

There are similar expressions for the other coefficients which appear in the self-consistent field equations (13) and (14).

The generalization of Eq. (81) takes the form

$$\begin{aligned} \dot{E}_1 &= \alpha_1 E_1 - \beta_1 E_1^3 - \theta_{12} E_1 E_2^2 - \theta_{13} E_1 E_3^2 - (\eta_{23} \cos \psi + \xi_{23} \sin \psi) E_2^2 E_3, \\ \dot{E}_2 &= \alpha_2 E_2 - \beta_2 E_2^3 - \theta_{21} E_2 E_1^2 - \theta_{23} E_2 E_3^2 - (\eta_{13} \cos \psi + \xi_{23} \sin \psi) E_1 E_2 E_3, \\ \dot{E}_3 &= \alpha_3 E_3 - \beta_3 E_3^3 - \theta_{31} E_3 E_1^2 - \theta_{32} E_3 E_2^2 - (\eta_{21} \cos \psi + \xi_{21} \sin \psi) E_2^2 E_1. \end{aligned} \quad (108)$$

The coefficients α_n and β_n were already calculated in the single-frequency case, and are given by Eqs. (82) and (83). The other coefficients are given by

$$\theta_{12} = \frac{1}{16} \pi^{1/2} \nu (\varphi^4 / (\epsilon_0 \hbar^3 K u)) \{ \bar{N} \gamma_{ab}^2 (\gamma_a \gamma_b)^{-1} [\mathfrak{L}(\omega - \nu_{12}) + 4\Delta^{-2}] - 2\bar{N} \Delta^{-2} + N_2 \Delta^{-2} [\gamma_{ab}^2 - (\omega - \nu_1) \Delta] \mathfrak{L}(\omega - \nu_1) \}, \quad (109)$$

$$\theta_{13} = \frac{1}{16} \pi^{1/2} \nu (\varphi^4 / (\epsilon_0 \hbar^3 K u)) \{ \bar{N} \gamma_{ab}^2 (\gamma_a \gamma_b)^{-1} [\mathfrak{L}(\omega - \nu_2) + \Delta^{-2}] - \frac{1}{2} \bar{N} \Delta^{-2} + \frac{1}{4} N_4 \Delta^{-2} [\gamma_{ab}^2 - 2(\omega - \nu_1) \Delta] \mathfrak{L}(\omega - \nu_1) \}, \quad (110)$$

$$\eta_{23} = \frac{1}{16} \pi^{1/2} \nu (\varphi^4 / (\epsilon_0 \hbar^3 K u \Delta^2)) \{ N_2 [\gamma_{ab}^2 - (\omega - \nu_{12}) \Delta] \mathfrak{L}(\omega - \nu_{12}) - \bar{N} \}, \quad (111)$$

$$\xi_{23} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u \Delta^2)^{-1} N_2 \gamma_{ab} (\omega - \nu_{12} + \Delta) \mathfrak{L}(\omega - \nu_{12}), \quad (112)$$

$$\theta_{21} = \frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} \gamma_{ab}^2 (\gamma_a \gamma_b)^{-1} [\mathfrak{L}(\omega - \nu_{12}) + 4\Delta^{-2}] + N_2 \Delta^{-2} [\gamma_{ab}^2 + (\omega - \nu_2) \Delta] \mathfrak{L}(\omega - \nu_2) - 2\bar{N} \Delta^{-2} \}, \quad (113)$$

$$\theta_{23} = \frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} \gamma_{ab}^2 (\gamma_a \gamma_b)^{-1} [\mathfrak{L}(\omega - \nu_2) + 4\Delta^{-2}] + N_2 \Delta^{-2} [\gamma_{ab}^2 - (\omega - \nu_2) \Delta] \mathfrak{L}(\omega - \nu_2) - 2\bar{N} \Delta^{-2} \}, \quad (114)$$

$$\xi_{13} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u \Delta^2)^{-1} N_2 \gamma_{ab} [(\omega - \nu_{23} - \Delta) \mathfrak{L}(\omega - \nu_{23}) + (\omega - \nu_{12} + \Delta) \mathfrak{L}(\omega - \nu_{12})], \quad (115)$$

$$\eta_{13} = \frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u \Delta^2)^{-1} \{ N_2 [\gamma_{ab}^2 + (\omega - \nu_{23}) \Delta] \mathfrak{L}(\omega - \nu_{23}) + N_2 [\gamma_{ab}^2 - (\omega - \nu_{12}) \Delta] \mathfrak{L}(\omega - \nu_{12}) + 2\bar{N} \}, \quad (116)$$

$$\theta_{31} = \frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} \gamma_{ab}^2 (\gamma_a \gamma_b)^{-1} [\mathfrak{L}(\omega - \nu_2) + \Delta^{-2}] + \frac{1}{4} N_4 \Delta^{-2} [\gamma_{ab}^2 + 2(\omega - \nu_3) \Delta] \mathfrak{L}(\omega - \nu_3) - \frac{1}{2} \bar{N} \Delta^{-2} \}, \quad (117)$$

$$\theta_{32} = \frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} \gamma_{ab}^2 (\gamma_a \gamma_b)^{-1} [\mathfrak{L}(\omega - \nu_{23}) + 4\Delta^{-2}] + N_2 \Delta^{-2} [\gamma_{ab}^2 + (\omega - \nu_3) \Delta] \mathfrak{L}(\omega - \nu_3) - 2\bar{N} \Delta^{-2} \}, \quad (118)$$

$$\eta_{21} = \frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u \Delta^2)^{-1} \{ N_2 [\gamma_{ab}^2 + (\omega - \nu_{23}) \Delta] \mathfrak{L}(\omega - \nu_{23}) - \bar{N} \}, \quad (119)$$

$$\xi_{21} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u \Delta^2)^{-1} N_2 \gamma_{ab} (\omega - \nu_{23} - \Delta) \mathfrak{L}(\omega - \nu_{23}). \quad (120)$$

The frequency and phase determining equations are of the form

$$\nu_1 + \dot{\phi}_1 = \Omega_1 + \sigma_1 + \rho_1 E_1^2 + \tau_{12} E_2^2 + \tau_{13} E_3^2 + E_2^2 E_3 E_1^{-1} (\eta_{23} \sin\psi - \xi_{23} \cos\psi), \quad (121)$$

$$\nu_2 + \dot{\phi}_2 = \Omega_2 + \sigma_2 + \rho_2 E_2^2 + \tau_{21} E_1^2 + \tau_{23} E_3^2 + E_1 E_3 (\eta_{13} \sin\psi - \xi_{13} \cos\psi), \quad (122)$$

$$\nu_3 + \dot{\phi}_3 = \Omega_3 + \sigma_3 + \rho_3 E_3^2 + \tau_{31} E_1^2 + \tau_{32} E_2^2 + E_2^2 E_1 E_3^{-1} (\eta_{21} \sin\psi - \xi_{21} \cos\psi), \quad (123)$$

where the coefficients σ_n and ρ_n are already given by Eqs. (90) and (91), and the η 's and ξ 's by Eqs. (111), (112), (115), (116), (119), and (120). The remaining coefficients are

$$\tau_{12} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} (\gamma_a \gamma_b)^{-1} [\gamma_{ab} (\omega - \nu_{12}) \mathcal{L}(\omega - \nu_{12}) + 2\gamma_{ab} \Delta^{-1}] + N_2 \Delta^{-2} \gamma_{ab} (\omega - \nu_1 + \Delta) \mathcal{L}(\omega - \nu_1) \}, \quad (124)$$

$$\tau_{13} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} (\gamma_a \gamma_b)^{-1} [\gamma_{ab} (\omega - \nu_{13}) \mathcal{L}(\omega - \nu_{13}) + \gamma_{ab} \Delta^{-1}] + \frac{1}{4} N_4 \Delta^{-2} \gamma_{ab} (\omega - \nu_1 + 2\Delta) \mathcal{L}(\omega - \nu_1) \}, \quad (125)$$

$$\tau_{21} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} (\gamma_a \gamma_b)^{-1} [\gamma_{ab} (\omega - \nu_{12}) \mathcal{L}(\omega - \nu_{12}) - 2\gamma_{ab} \Delta^{-1}] + N_2 \Delta^{-2} \gamma_{ab} (\omega - \nu_2 - \Delta) \mathcal{L}(\omega - \nu_2) \}, \quad (126)$$

$$\tau_{23} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} (\gamma_a \gamma_b)^{-1} [\gamma_{ab} (\omega - \nu_{23}) \mathcal{L}(\omega - \nu_{23}) + 2\gamma_{ab} \Delta^{-1}] + N_2 \Delta^{-2} \gamma_{ab} (\omega - \nu_2 + \Delta) \mathcal{L}(\omega - \nu_2) \}, \quad (127)$$

$$\tau_{31} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} (\gamma_a \gamma_b)^{-1} [\gamma_{ab} (\omega - \nu_{12}) \mathcal{L}(\omega - \nu_{12}) - \gamma_{ab} \Delta^{-1}] + \frac{1}{4} N_4 \Delta^{-2} \gamma_{ab} (\omega - \nu_3 - 2\Delta) \mathcal{L}(\omega - \nu_3) \}, \quad (128)$$

$$\tau_{32} = -\frac{1}{16} \pi^{1/2} \nu \varphi^4 (\epsilon_0 \hbar^3 K u)^{-1} \{ \bar{N} (\gamma_a \gamma_b)^{-1} [\gamma_{ab} (\omega - \nu_{23}) \mathcal{L}(\omega - \nu_{23}) - 2\gamma_{ab} \Delta^{-1}] + N_2 \Delta^{-2} \gamma_{ab} (\omega - \nu_3 - \Delta) \mathcal{L}(\omega - \nu_3) \}. \quad (129)$$

10. TWO-FREQUENCY OPERATION

We may here drop all terms referring to the third frequency ν_3 . There are now no "combination tones" in near resonance with Ω_1 and Ω_2 . The amplitudes E_1 and E_2 are determined by the differential equations

$$\begin{aligned} \dot{E}_1 &= \alpha_1 E_1 - \beta_1 E_1^3 - \theta_{12} E_1 E_2^2, \\ \dot{E}_2 &= \alpha_2 E_2 - \theta_{21} E_2 E_1^2 - \beta_2 E_2^3, \end{aligned} \quad (130)$$

where the coefficients α_n , β_n , θ_{12} , and θ_{21} are now given by Eqs. (82), (83), (109), and (113).

Introducing the squared amplitudes

$$X = E_1^2 \quad \text{and} \quad Y = E_2^2, \quad (131)$$

Eqs. (126) become

$$\begin{aligned} \dot{X} &= 2X(\alpha_1 - \beta_1 X - \theta_{12} Y), \\ \dot{Y} &= 2Y(\alpha_2 - \theta_{21} X - \beta_2 Y). \end{aligned} \quad (132)$$

The condition for a steady state of oscillation is $\dot{X}=0$, $\dot{Y}=0$ and may represent graphically in an X - Y plane by the point of intersection of the two straight lines

$$\begin{aligned} L_1: \quad & \beta_1 X + \theta_{12} Y = \alpha_1, \\ L_2: \quad & \theta_{21} X + \beta_2 Y = \alpha_2, \end{aligned} \quad (133)$$

if there is one in the first quadrant, together with the single-frequency solutions

$$X = \alpha_1/\beta_1, \quad Y = 0 \quad \text{and} \quad X = 0, \quad Y = \alpha_2/\beta_2. \quad (134)$$

The differential equations (132) allow us to follow the temporal behavior of the state X , Y of oscillation in the phase plane X , Y . Through any point in this plane (except stationary points) there passes a curve which indicates the path followed by the representative point (X, Y) on its way to a stable state of oscillation. The parametric equations of the curves are $X = X(t)$, $Y = Y(t)$ with the time t as parameter. From the differential

equations it is seen that each curve has a vertical tangent when it crosses the first of the straight lines (133), and a horizontal tangent when it crosses the second straight line. This principle facilitates a very simple, if qualitative, graphical integration of the differential equations (132) for the paths of the phase points in the cases discussed below.

The various possibilities are depicted¹⁸ in Figs. 3-5 where it is assumed that both α_1 and α_2 are positive,

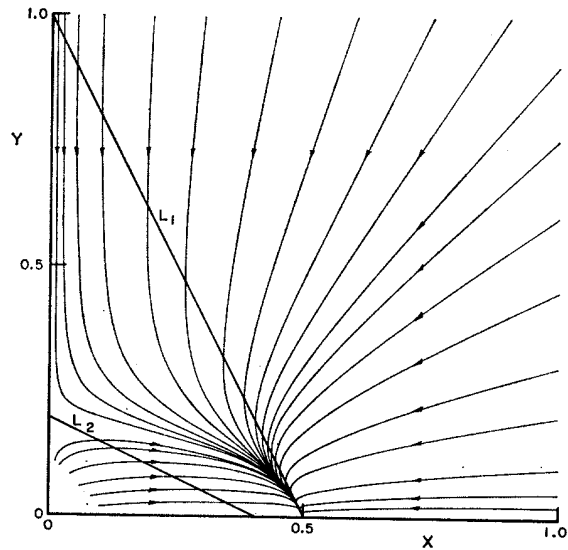


FIG. 3. Phase curves showing the transient behavior of two-mode oscillation. The straight lines L_1 and L_2 of Eq. (133) are taken to have coefficients $\alpha_1 = 1$, $\alpha_2 = 0.4$, $\beta_1 = \beta_2 = 2$, $\theta_{12} = \theta_{21} = 1$. The slope of a phase curve is zero when it crosses line L_2 and infinite when it crosses line L_1 . Although both modes are above threshold, the favored X oscillation is able to quench the Y oscillation.

¹⁸ The phase paths of Figs. 2-4 were kindly integrated on an analog computer by Dr. B. Wise of the Engineering Science Laboratory, Oxford, to whom the author is very indebted.

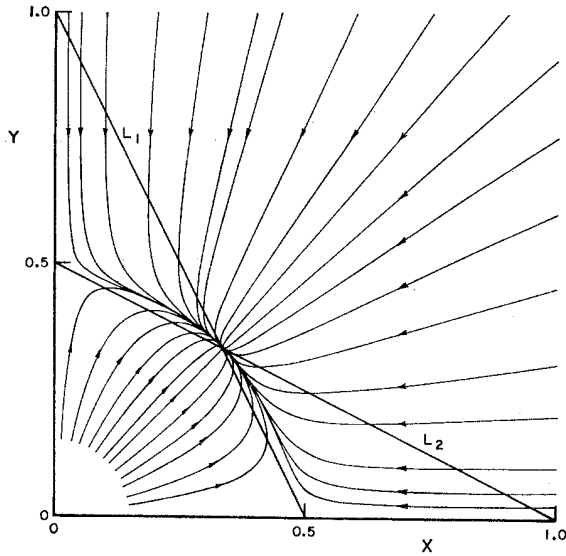


FIG. 4. Diagram similar to Fig. 3, except that the gain parameter for the second mode has been raised to $\alpha_2=1$. Simultaneous oscillations at both frequencies occur at the single stable steady state. Both Figs. 3 and 4 correspond to "weak" coupling.

so that the two modes are individually above threshold. The coefficients β_n are necessarily positive if the optical medium is an active one, and while the mode competition coefficients θ could conceivably have the opposite sign, they have been assumed positive (and equal to each other) in drawing the figures.

Figure 3 applies when mode 1 is well above threshold, but mode 2 is only a little above its threshold, either because the cavity resonance frequency Ω_2 is detuned from the atomic transition frequency ω or possibly because $Q_2 < Q_1$. It is clear that the point $(\alpha_1/\beta_1, 0)$ represents a stable state of oscillation, while $(0, \alpha_2/\beta_2)$ corresponds to an unstable steady state. Hence there is a range of operation above threshold of modes 1 and 2 where oscillations in the favored mode 1 are able to inhibit oscillations at the second frequency. One might say that the effective gain for the second mode

$$\alpha_2' = \alpha_2 - \theta X = \alpha_2 - \theta \alpha_1 / \beta_1$$

is being made negative by the presence of oscillations at ν_1 . From Eqs. (109) and (113) it is seen that the inhibiting effect is enhanced when $\nu_{12} = \omega$, or when the two cavity modes are on opposite sides of the atomic transition frequency, and approximately equally far from it. The physical interpretation of this effect which clearly involves o.f. saturation will be brought out more clearly in Sec. 18.

As the excitation increases, α_2' will eventually become positive, and the relevant diagrams are Figs. 4 and 5. The former applies when $\beta_1\beta_2 > \theta^2$ (weak coupling) and the latter when $\beta_1\beta_2 < \theta^2$ (strong coupling).

The two cases of weak and strong coupling give very different behaviors. For weak coupling the point of

intersection of the two straight lines gives stable steady-state operation, while the single-frequency operating points are unstable. The optical maser oscillates simultaneously at two frequencies under these conditions. For strong coupling, on the other hand, the point of intersection of the two straight lines represents an unstable steady state and would not be realized in practice. All other points in the state diagram evolve into one or the other of the single-frequency operation points. Which of the two is reached depends on the past history of the state of oscillation. In other words, there is hysteresis.

In the Doppler broadened gaseous optical maser Eqs. (83), (109), and (113) indicate that the case of weak coupling is naturally favored, since $\beta_1\beta_2$ tends to be greater than $\theta_{12}\theta_{21}$. Hence, with possible exceptions such as the one discussed in the next section, double frequency operation is preferred. However, when van der Pol's theory of a double resonance feed back triode oscillator is transcribed into our notation, one finds that in his case $\theta = 2\beta$. This results from his assumption of a term $(E_1 \cos \nu_1 t + E_2 \cos \nu_2 t)^3$ in the triode output current. After discarding terms which have frequencies far from ν_1 and ν_2 , this becomes

$$\frac{3}{4}(E_1^3 + 2E_1E_2^2) \cos \nu_1 t + \frac{3}{4}(E_2^3 + 2E_2E_1^2) \cos \nu_2 t,$$

which leads to the stated relation $\theta = 2\beta$. The van der Pol oscillator prefers operation at a single frequency and exhibits hysteresis phenomena, as in the case of strong coupling. Evidently the atomic medium of an optical

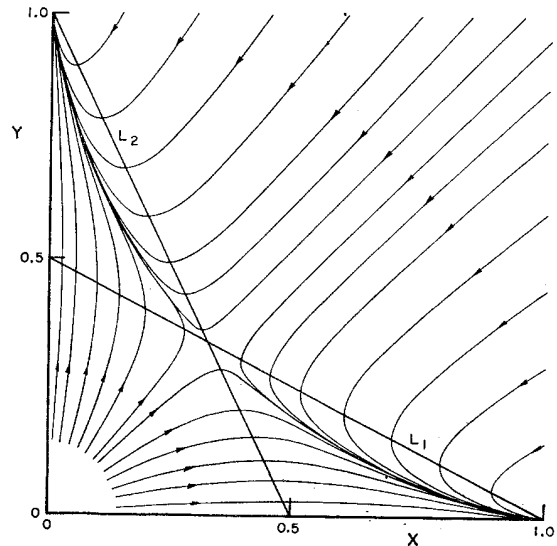


FIG. 5. Phase curves showing the transient behavior of two-mode oscillation when the straight lines L_1 and L_2 of Eq. (133) are taken to have the coefficients $\alpha_1 = \alpha_2 = 1$, $\beta_1 = \beta_2 = 1$, $\theta_{12} = \theta_{21} = 2$ (strong coupling). There are two possible stable steady states, each corresponding to single-frequency operation. The particular state reached depends on the initial conditions. Hysteresis phenomena would occur if the parameters characterizing the oscillator were slowly changed.

maser differs from van der Pol's triode oscillator because the nonlinear response of the atoms to frequencies ν_1 and ν_2 has a resonant character not assumed of the triode. It will become clearer from the discussion of Sec. 18 that two groups of atoms with different velocity are driving the two oscillations with only a limited degree of interference. One could easily make a comparable model of the van der Pol type with *two* triodes each with its own tank circuit. A small amount of coupling between the two oscillators would produce intermode effects described by the θ coefficients in the weak-coupling case.

There is also a further case in which the two straight lines coincide, i.e., when $\alpha_1/\alpha_2 = \beta_1/\theta = \theta/\beta_2$. In that case, there is a neutral steady state for the representative point lying anywhere on the line within the first quadrant. In practice, the state of operation when this condition is nearly satisfied should be very sensitive to microphonic disturbances.

11. INTENSITIES AND FREQUENCIES IN TWO-MODE OPERATION

The two-mode steady-state solution of (130) is given by

$$\begin{aligned} E_1^2 &= (\beta_2\alpha_1 - \theta_{12}\alpha_2) / (\beta_1\beta_2 - \theta_{12}\theta_{21}), \\ E_2^2 &= (\beta_1\alpha_2 - \theta_{21}\alpha_1) / (\beta_1\beta_2 - \theta_{12}\theta_{21}), \end{aligned} \quad (135)$$

where, as explained before, the right-hand sides may be evaluated for $\nu_1 = \Omega_1$ and $\nu_2 = \Omega_2$ without appreciable error. The frequencies are obtained by dropping inapplicable terms from Eqs. (121)–(123). We may set $\dot{\phi}_1 = \dot{\phi}_2$ without loss of generality, and find

$$\begin{aligned} \nu_1 &= \Omega_1 + \sigma_1 + \rho_1 E_1^2 + \tau_{12} E_2^2, \\ \nu_2 &= \Omega_2 + \sigma_2 + \rho_2 E_2^2 + \tau_{21} E_1^2, \end{aligned} \quad (136)$$

where the right-hand sides are to be evaluated for $\nu_n = \Omega_n$, and the E_n^2 are as given in (135).

These equations are fairly complicated, and probably can be used in full generality only for a numerical analysis¹⁹ of very detailed data on optical maser operation. Such a study would be simplified if the values of the E 's could be inferred experimentally, since that would effectively reduce the dependence of Eqs. (136) on cavity tuning. For the present we will merely work out the frequencies for the important case of "mid-tuning" where $\nu_{12} = \frac{1}{2}(\nu_1 + \nu_2) = \omega$, $\omega - \nu_1 = \frac{1}{2}\Delta$ and $\nu_2 - \omega = \frac{1}{2}\Delta$. If we regard Δ as being much greater than the γ 's, we then may approximate the coefficients appearing in Eqs. (135)–(136) as follows:

$$\beta_1 \approx \beta_2 \approx \theta_{12} \approx \theta_{21} \approx \frac{1}{16} \pi^{1/2} \wp^4 \bar{N} (\epsilon_0 \hbar^3 K u \gamma_a \gamma_b)^{-1} = \beta, \quad (137)$$

$$\rho_2 \approx -\rho_1 = 2\gamma_{ab} \Delta^{-1} \beta = \rho, \quad (138)$$

$$\tau_{12} \approx -\tau_{21} \approx -\rho, \quad (139)$$

and assuming $Q_1 = Q_2 = Q$, the gain parameters become

$$\alpha_1 \approx \alpha_2 \approx \frac{1}{2} \nu Q_n^{-1} \{ \Im [Z_i(\frac{1}{2}\Delta) / Z_i(0)] - 1 \} = \alpha. \quad (140)$$

The intensities are $E_1^2 = E_2^2 = \frac{1}{2} \alpha / \beta$, and the frequencies are

$$\begin{aligned} \nu_1 &= \Omega_1 + \sigma_1 - \rho \alpha / \beta, \\ \nu_2 &= \Omega_2 + \sigma_2 + \rho \alpha / \beta, \end{aligned} \quad (141)$$

so that the beat frequency is

$$\nu_2 - \nu_1 \approx \Delta + (\sigma_2 - \sigma_1) + 4\gamma_{ab} \alpha / \Delta. \quad (142)$$

It will be seen that in the approximations of Eq. (137) we have $\beta_1 \beta_2 = \theta_{12} \theta_{21}$, and hence in order to decide whether the coupling is "weak" or "strong" it is necessary to evaluate the coefficients β and θ more exactly. In case of exact midtuning, one finds that $\beta^2 - \theta^2$ has the same sign as $\bar{N} + N_2$. Ordinarily this would be positive since \bar{N} must be positive for oscillations to occur. However, it is possible in principle to arrange to have $\bar{N} > 0$ and $N_2 < -\bar{N}$ by having $N(z) > 0$ in the middle two quarters of the tube length, and $N(z) < 0$ in the end quarters. In practice the last requirement could be met by adjusting the gas discharge conditions near the ends so that the lower maser atomic level is more populated than the upper one. In He-Ne masers an increase of tube diameter near the ends might be helpful in this respect. Diffusion of Ne metastables to the walls is thereby reduced, and electron excitation of the lower maser level is increased.

12. NORMAL THREE-FREQUENCY OPERATION

Equations (108) and (121)–(123) are fairly complicated, but can be readily used to discuss a number of special cases, as indicated in the following three sections.

In general, unless care is taken to adjust the cavity tuning very accurately, the three frequencies ν_1 , ν_2 , and ν_3 will be such that the relative phase angle ψ of Eq. (106) is a linear function of the time. Then the last terms in Eqs. (108), (121)–(123) are periodic functions of time, and in some approximation their effects average out. If we neglect these terms, we can get a steady-state solution for the intensities E_1^2 , E_2^2 , E_3^2 from the system of inhomogeneous linear equations

$$\begin{aligned} \alpha_1 &= \beta_1 E_1^2 + \theta_{12} E_2^2 + \theta_{13} E_3^2, \\ \alpha_2 &= \beta_2 E_2^2 + \theta_{21} E_1^2 + \theta_{23} E_3^2, \\ \alpha_3 &= \beta_3 E_3^2 + \theta_{31} E_1^2 + \theta_{32} E_2^2, \end{aligned} \quad (143)$$

and for the frequencies ν_1 , ν_2 , ν_3 from

$$\begin{aligned} \nu_1 &= \Omega_1 + \sigma_1 + \rho_1 E_1^2 + \tau_{12} E_2^2 + \tau_{13} E_3^2, \\ \nu_2 &= \Omega_2 + \sigma_2 + \rho_2 E_2^2 + \tau_{21} E_1^2 + \tau_{23} E_3^2, \\ \nu_3 &= \Omega_3 + \sigma_3 + \rho_3 E_3^2 + \tau_{31} E_1^2 + \tau_{32} E_2^2, \end{aligned} \quad (144)$$

again taking $\dot{\phi}_1 = \dot{\phi}_2 = \dot{\phi}_3 = 0$. Since the coefficients in Eqs. (143) and (144) are slowly varying functions of frequency, it will suffice first to determine the E_n^2 from

¹⁹ Such an analysis is being made by Dr. R. L. Fork and Dr. M. A. Pollack. The author is very grateful to them for helpful discussions on this and other parts of the manuscript.

(143) and then to calculate the ν_n 's from (144). The equations are a fairly obvious generalization of those for the two frequency case. It will be noted that operation with $E_1^2 \neq 0$, $E_2^2 \neq 0$ can inhibit normal oscillation at ν_3 until, with increasing excitation,

$$\alpha_3' = \alpha_3 - \theta_{31}E_1^2 - \theta_{32}E_2^2 \quad (145)$$

becomes positive.

Further discussion of pulling, pushing, and mode competition for this case will be left to readers requiring a numerical analysis of their data.

13. COMBINATION TONES

As mentioned in Secs. 7, 9, the third-order polarization $P_n^{(3)}(t)$ of the active medium has constituents which oscillate at all possible frequencies of the form $\nu_\mu - \nu_\rho + \nu_\sigma$. Even for "two"-frequency oscillation, there are additional frequencies $2\nu_2 - \nu_1 \equiv \nu_3'$ and $2\nu_1 - \nu_2 \equiv \nu_0'$ in the polarization which are very close to resonance with the principal cavity modes just above and below the two Ω_1 and Ω_2 of main interest. As a consequence of Maxwell's equations, fields at frequencies ν_3' and ν_0' necessarily exist in the cavity and can appear in the output. Well below the threshold for normal three-frequency oscillation, E_3 (and also E_0) will be much smaller than E_1 and E_2 . We also assume $\gamma_{ab} \ll \Delta \ll Ku$ and $\nu_2 \simeq \omega$ so that

$$\eta_{21} \simeq -\frac{1}{16}\pi^{1/2}\nu\varphi^4(\epsilon_0\hbar^3Ku\Delta^2)^{-1}[\bar{N} + 2N_2], \quad (146)$$

$$\xi_{21} \simeq 0.$$

For a steady state, the third Eq. (108) then gives

$$E_3 \simeq \eta_{21}E_2^2E_1[\alpha_3 - \theta_{31}E_1^2 - \theta_{32}E_2^2]^{-1} \cos\psi. \quad (147)$$

The relative phase angle ψ is determined by Eq. (123) to have $\sin\psi \simeq 0$, whence $|\cos\psi| \simeq 1$, and

$$E_3 \simeq -\frac{1}{16}\pi^{1/2}\nu\varphi^4(\epsilon_0\hbar^3Ku\Delta^2)^{-1} \times [\alpha_3 - \theta_{31}E_1^2 - \theta_{32}E_2^2]^{-1} |\bar{N} + 2N_2| E_2^2 E_1. \quad (148)$$

This expression is intended to be used under excitation conditions for which the denominator is negative. When the excitation increases, and the factor involving α_3 turns positive, the neglected nonlinear terms in E_3 would have to be taken into account in order to describe the previously discussed normal three-frequency operation.

In order to obtain the combination tone ($2\nu_2 - \nu_1$) experimentally, one should adjust the cavity tuning so Ω_2 is slightly above the atomic transition frequency ω , thereby making Ω_1 a little nearer resonance than Ω_3 . The excitation should be increased until "two"-frequency operation is obtained, but not yet genuine three-frequency operation.

Under these conditions, the θ 's in Eq. (148) contain a factor Δ^{-2} and to simplify the discussion they will now be neglected. Equation (148) then has a factor in its denominator

$$\alpha_3 \simeq -\frac{1}{2}(\nu/Q_3)/G_3, \quad (149)$$

where the gain factor G_3 is given approximately by

$$G_3 \simeq [1 - (\bar{N}/\bar{N}_T)(Z_i(\Delta)/Z_i(0))]^{-1}, \quad (150)$$

if one is not too near to threshold for normal three-frequency operation. Equations (148)–(150), within their domain of validity, indicate that E_3 is smaller than E_1 by a factor

$$E_3/E_1 \simeq \frac{1}{8}\pi^{1/2}\varphi^4E_2^2(\epsilon_0\hbar^3Ku\Delta^2)^{-1}Q_3G_3|\bar{N} + 2N_2|. \quad (151)$$

Using Eq. (62) for $n=3$, this can be expressed in a convenient form

$$E_3/E_1 = \frac{1}{4}[\frac{1}{2}(\varphi E_2)^2/(\hbar^2\gamma_a\gamma_b)] [(\gamma_a\gamma_b)/\Delta^2] G_3 \times |1 + (2N_2/\bar{N})| \exp(\Delta/Ku)^2, \quad (152)$$

which shows how the amplitude of the combination tone depends on G_3 , and on the saturation parameter as given by Eq. (96) for $n=2$. It should be noted that Eq. (152) could vanish if the spatial distribution of the excitation density is such that $2N_2 + \bar{N} = 0$. (If the excitation is confined to the central region of the Fabry-Perot tube, N_2 and \bar{N} , by Eqs. (47), (58) have opposite signs.) An experimental study of the above phenomena might facilitate determination of some of the quantities which enter into our equations but for which direct experimental values are not yet available.

14. FREQUENCY LOCKING PHENOMENA

It has been observed by Javan²⁰ and by Fork¹⁹ that when the cavity tuning is gradually changed in normal three-frequency operation so that the separation of the beat notes $\nu_2 - \nu_1$ and $\nu_3 - \nu_2$ approaches a small value (typically of order 1 kc/sec), a frequency jump occurs. This phenomenon can be easily understood by reference to Eqs. (121)–(123). For simplicity, we neglect the small frequency pushing associated with the terms involving ρ_{nm} and τ_{nm} , since the nonlinear pulling terms σ_n already give sufficient generality to the frequency relationships. By subtracting the sum of Eqs. (121)–(123) from twice Eq. (122), we find a differential equation for the relative phase angle ψ of Eq. (106) in the form

$$\dot{\psi} = \sigma + A \sin\psi + B \cos\psi, \quad (153)$$

where

$$\sigma = 2\sigma_2 - \sigma_1 - \sigma_3, \quad (154)$$

and A and B are slowly varying quantities which depend on the E_n , ξ_{nm} , and η_{nm} . We evaluate the ξ_{nm} and η_{nm} with the usual approximations $\gamma_{ab} \ll \Delta \ll Ku$. Then

$$\eta_{23} \simeq \eta_{21} \simeq -\frac{1}{16}\pi^{1/2}\nu\varphi^4(\epsilon_0\hbar^3Ku\Delta^2)^{-1}\{\bar{N} + 2N_2\},$$

$$\eta_{13} \simeq \frac{1}{8}\pi^{1/2}\nu\varphi^4(\epsilon_0\hbar^3Ku\Delta^2)^{-1}\{\bar{N} - 2N_2\}, \quad (155)$$

$$\xi_{23} \simeq \xi_{21} \simeq \xi_{13} \simeq 0.$$

Because of the nearly symmetrical tuning of ν_1 and ν_3

²⁰ A. Javan, private communications for which the author is very grateful.

we may set $E_1 = E_3$ and find

$$A \approx \frac{1}{8}\pi^{1/2}\nu \wp^4 (\epsilon_0 \hbar^3 K u \Delta^2)^{-1} \times \{(\bar{N} + 2N_2)E_2^2 + 2(\bar{N} - 2N_2)E_1^2\}, \quad (156)$$

$B \approx 0$.

The differential equation

$$\dot{\psi} = \sigma + A \sin\psi + B \cos\psi \quad (157)$$

has an implicit solution

$$t(\psi) = \int_{\psi_0}^{\psi} dx / (\sigma + A \sin x + B \cos x), \quad (158)$$

where ψ_0 is the value of ψ at $t=0$. The character of the function $\psi(t)$ depends critically on whether

$$(A^2 + B^2)^{1/2} < |\sigma| \quad \text{or} \quad (A^2 + B^2)^{1/2} > |\sigma|.$$

In the first case, the integrand of (158) has no singularities in the range of integration, and one finds that as ψ approaches infinity so does t . Asymptotically, apart from pulsations, one has $\psi \approx \sigma t + \text{const}$. On insertion of this value in Eqs. (121)–(123) one finds that the frequencies are given by

$$\nu_n \sim \Omega_n + \sigma_n \quad (159)$$

apart from pushing effects and pulsations of phase φ_n , in agreement with the results of Sec. 6 with the approximations made here.

In the second case, $(A^2 + B^2) > \sigma^2$, the integral diverges, i.e., $t \rightarrow \infty$ when ψ reaches the value $-\sin^{-1} \times (\sigma / (A^2 + B^2)^{1/2})$. In other words, $\psi(t)$ approaches this value asymptotically. The disappearance from ψ of any linear dependence on t forces $\nu_3 - \nu_2 = \nu_2 - \nu_1$ and frequency locking ensues, with a definite relative phase angle $2\varphi_2 - \varphi_1 - \varphi_3$.

Let us suppose that the maser is in normal three-frequency operation with two distinct beat notes $\nu_3 - \nu_2$ and $\nu_2 - \nu_1$ near to $\Delta \approx 150$ Mc/sec, i.e., $\sigma = (\nu_3 - \nu_2) - (\nu_2 - \nu_1)$ is somewhat greater than $(A^2 + B^2)^{1/2}$. As the middle cavity frequency Ω_2 is tuned closer to the atomic resonance frequency ω , the separation of beat note frequencies $|\sigma|$ decreases. There should be some pulsations in phase which would increase in amplitude as symmetrical tuning is approached. When $|\sigma|$ reaches $(A^2 + B^2)^{1/2}$, a quick transition to the locked state should be made, and only one beat note should be observed. Under the additional simplifying assumption $E_2 \gg E_1 \approx E_3$, and with use of the starting condition (60) for single-frequency oscillation, the separation of the two beat notes which could be attained just before locking occurs should be given by

$$|\sigma| = \frac{1}{8} [(\bar{N} + 2N_2) / \bar{N}_T] (\wp E_2 / \hbar \Delta)^2 \nu / Q, \quad (160)$$

which is conveniently expressed as a small fraction of the cavity bandwidth.

It might be pointed out that the above phenomenon

is very closely related to one discussed by van der Pol²¹ in 1924–27. He considered a self-sustained triode oscillator, capable of oscillation at frequency ν_1 . If an external signal at ν is injected into the tank circuit, it may be possible to detect a beat note at $|\nu - \nu_1|$ using a square-law detector. If, however, ν is tuned gradually towards ν_1 , a very sudden jump occurs, after which oscillations occur only at ν and the beat note disappears. The width of the “quiet” frequency range depends approximately linearly on the amplitude of the injected signal, when this is small. In the case of the optical maser, we can think that an oscillator at ν_1 is being perturbed by an “external” signal at the combination tone frequency $\nu = 2\nu_2 - \nu_3$ which arises from the third-order polarization $P_1^{(3)}(t)$ induced in the nonlinear active medium.

15. POPULATION CHANGES AND PULSATIONS

In the absence of o.f. oscillations, the density of atoms in one of the two maser states, say a , can be determined by suitable integrations from $\rho_{aa}^{(0)}(a, z_0, t_0, \nu, t)$ as given by Eq. (24). When oscillations set in, there are contributions of second order which can be calculated from $\rho_{aa}^{(2)}(\alpha, z_0, t_0, \nu, t)$ using Eq. (31) for $\alpha = a$ and a similar equation for $\alpha = b$. One has

$$\rho_{aa}(z, t) = \int_{-\infty}^{\infty} d\nu \int_{-\infty}^t dt_0 \int dz_0 \delta(z - z_0 - \nu t + \nu t_0) \times \sum_{\alpha=a,b} \lambda_{\alpha}(z_0, t_0, \nu) \rho_{aa}(\alpha, z_0, t_0, \nu, t). \quad (161)$$

It will suffice merely to give the result. One finds, with obvious approximations

$$\begin{aligned} \rho_{aa}(z, t) = & [\Lambda_a(z, t) / \gamma_a] + \{[\Lambda_a(z, t) / \gamma_a] - [\Lambda_b(z, t) / \gamma_b]\} \\ & \times \frac{1}{8} (\wp^2 / K u) \sum_{\mu} \sum_{\rho} \{E_{\mu} E_{\rho} \mathfrak{D}_{\alpha}(\nu_{\mu} - \nu_{\rho}) i Z(\omega - \nu_{\rho}) \\ & \times \exp i[(\nu_{\mu} - \nu_{\rho})t + (\varphi_{\mu} - \varphi_{\rho})] + \text{c.c.}\} \\ & \times \cos[(n_{\mu} - n_{\rho})\pi z / L] \quad (162) \end{aligned}$$

apart from terms with rapid spatial oscillations.

For single-frequency operation, one finds

$$\begin{aligned} \rho_{aa}(z, t) = & [\Lambda_a(z, t) / \gamma_a] \\ & - \{[\Lambda_a(z, t) / \gamma_a] - [\Lambda_b(z, t) / \gamma_b]\} \\ & \times \frac{1}{4} (\wp E_1)^2 (\gamma_a K u)^{-1} Z_i(\omega - \nu_1), \quad (163) \end{aligned}$$

which contains the lowest order effects of o.f. saturation. In some cases, the density could be monitored by observation of the decay radiation emitted from state a (or b) in transition to some lower level. (Of course, if trapping of resonance radiation were involved, the interpretation would be somewhat complicated.) The change produced by o.f. radiation in $\rho_{aa}(z, t)$ might serve to aid in the determination of parameters like

²¹ B. van der Pol, Phil. Mag. 3, 65 (1927) and the review article cited in Ref. 17.

γ_a, E_1^2 , etc., which enter into all our equations, but which might not otherwise be known.

For two-frequency operation of the optical maser, ρ_{aa} has a pulsating constituent at a frequency near Δ besides a dc part. If we assume $\gamma_{ab} \sim \gamma_a \gg \Delta \ll Ku$

$$\begin{aligned} \rho_{aa}(z,t) &= [\Lambda_a(z,t)/\gamma_a] - \{[\Lambda_a(z,t)/\gamma_a] - [\Lambda_b(z,t)/\gamma_b]\} \\ &\quad \times \frac{1}{4}\pi^{1/2} \varphi^2 (Ku\gamma_a)^{-1} [E_1^2 \exp - (\omega - \nu_1)^2 / (Ku)^2 \\ &\quad + E_2^2 \exp - (\omega - \nu_2)^2 / (Ku)^2] + \{[\Lambda_a/\gamma_a] - [\Lambda_b/\gamma_b]\} \\ &\quad \times \frac{1}{4}\pi^{1/2} \varphi^2 (Ku\Delta)^{-1} E_1 E_2 [\exp - (\omega - \nu_1)^2 / (Ku)^2 \\ &\quad + \exp - (\omega - \nu_2)^2 / (Ku)^2] \cos(\pi z/L) \sin(\nu_2 - \nu_1)t. \end{aligned} \quad (164)$$

From this expression one sees that the amplitude of the pulsations at frequency near Δ , relative to the dc change in population due to o.f. oscillation, should be

$$2E_1 E_2 (E_1^2 + E_2^2)^{-1} (\gamma_a/\Delta) \cos(\pi z/L),$$

if, for simplicity, we neglect the Gaussian exponentials. Here again, it might be useful to use this phenomenon as a diagnostic tool while undertaking a systematic study of two-frequency operation.

16. CONNECTIONS WITH PREVIOUS CALCULATIONS

The basic paper in this field is, of course, that of Schawlow and Townes¹ who give expressions for threshold equivalent to (62). Townes²² has also given an equation for linear pulling, as has Javan²³ for nonlinear pulling.

Oscillations of an optical maser involve the propagation of radiation in a nonlinear medium. Several papers have recently appeared which deal with this subject. For various reasons, these do not apply very closely to our particular problem. Thus, Bloembergen *et al.*²⁴ and Franken and Ward²⁵ treated harmonic generation which plays a relatively minor role for us. Teng and Statz²⁶ discussed low-order nonlinearities in a gaseous medium, but, as will be discussed below, their treatment of Doppler broadening is not adequate for our purposes. Also our model for radiation damping is more realistic than theirs which involves just one relaxation time τ , while our equations contain two decay constants γ_a and γ_b . To be sure, the combination $\gamma_{ab} = \frac{1}{2}(\gamma_a + \gamma_b)$ enters most equations, and this might be identified with τ^{-1} .

Among other publications which deal with maser theory are those of Wagner and Birnbaum²⁷ and of

McCumber.²⁸ These papers consider to some extent the quantum nature of the electromagnetic field. They differ greatly in spirit and content from ours, and we will not attempt to make a comparison here. The work of Haken and Sauerermann²⁹ and of Davis³⁰ seems much closer to ours, but there are significant differences in the models used, and in the appearance of our equations.

As mentioned in Sec. 4 an earlier calculation⁸ applicable to an optical gaseous maser neglected complications arising from the atomic motions and multimode oscillation. It was then possible to work with a density matrix $\rho(\mathbf{r},t)$ characterizing an ensemble⁷ of atoms at position \mathbf{r} at time t which were excited at any time $t_0 \leq t$. This obeyed

$$i\dot{\rho} = [\mathcal{H}, \rho] - (i/2)(\Gamma\rho + \rho\Gamma) + i\Lambda, \quad (165)$$

which differs from (18) by the term containing a source matrix Λ describing the (slowly varying) rate densities of excitations Λ_a and Λ_b

$$\Lambda = \begin{pmatrix} \Lambda_a & 0 \\ 0 & \Lambda_b \end{pmatrix}. \quad (166)$$

(In most applications Λ will be a diagonal matrix.)

It is possible to carry the calculation to higher order in the E_n for multimode oscillation without atomic motion by an iterative procedure in which we begin by neglecting any time dependence in the population difference $\rho_{aa} - \rho_{bb}$. In the rotating wave approximation, one of Eqs. (25) then gives a quasisteady-state solution for $\rho_{ab}(z,t)$

$$\begin{aligned} \rho_{ab}(z,t) &= -\frac{1}{2}i(\varphi/\hbar) \sum_{\mu} E_{\mu} U_{\mu}(z) \mathcal{D}(\omega - \nu_{\mu}) \\ &\quad \times (\rho_{aa} - \rho_{bb}) \exp - i(\nu_{\mu}t + \varphi_{\mu}). \end{aligned} \quad (167)$$

Inserting this in another one of Eqs. (25) and again making a rotating wave approximation we find rate equations

$$\begin{aligned} \dot{\rho}_{aa} &= -\gamma_a \rho_{aa} + R(\rho_{bb} - \rho_{aa}) + \Lambda_a, \\ \dot{\rho}_{bb} &= -\gamma_b \rho_{bb} + R(\rho_{aa} - \rho_{bb}) + \Lambda_b, \end{aligned} \quad (168)$$

where

$$\begin{aligned} R &= \frac{1}{4}(\varphi/\hbar)^2 \sum_{\lambda} \sum_{\mu} E_{\lambda} E_{\mu} U_{\lambda}(z) U_{\mu}(z) \\ &\quad \times [\mathcal{D}(\omega - \nu_{\mu})(\exp i(\nu_{\lambda} - \nu_{\mu})t + i(\varphi_{\lambda} - \varphi_{\mu})) + \text{c.c.}]. \end{aligned} \quad (169)$$

The "rate constant" R has pulsations for cases of multi-frequency operation, and through Eq. (168) these would lead to pulsations in the populations ρ_{aa} and ρ_{bb} at all beat frequencies $\nu_{\lambda} - \nu_{\mu}$. If it were deemed necessary to continue the iteration procedure the pulsating population difference $\rho_{aa} - \rho_{bb}$ could be approximately evalu-

²² C. H. Townes, *Advances in Quantum Electronics*, edited by J. Singer (Columbia University Press, New York, 1961), pp. 3-11.

²³ A. Javan, E. A. Ballik, and W. L. Bond, *J. Opt. Soc. Am.* **52**, 96 (1962).

²⁴ J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, *Phys. Rev.* **127**, 1918 (1962).

²⁵ P. A. Franken and J. F. Ward, *Rev. Mod. Phys.* **35**, 23 (1963).

²⁶ C. L. Tang and H. Statz, *Phys. Rev.* **128**, 1013 (1962).

²⁷ W. G. Wagner and G. Birnbaum, *J. Appl. Phys.* **32**, 1185 (1961).

²⁸ D. E. McCumber, *Phys. Rev.* **130**, 675 (1963).

²⁹ H. Haken and H. Sauerermann, *Z. Physik* **173**, 261 (1963); **176**, 47 (1963).

³⁰ L. W. Davis, *Proc. Inst. Elec. Engrs.* **51**, 76 (1963).

ated from Eq. (168) and put in Eqs. (25) to obtain an improved Eq. (167).

If we neglect the pulsations in R , we obtain

$$R = \frac{1}{2} (\varphi/\hbar)^2 \gamma_{ab} \sum_{\mu} E_{\mu}^2 [U_{\mu}(z)]^2 \mathcal{L}(\omega - \nu_{\mu}), \quad (170)$$

which is a plausible generalization of the rate of transitions previously obtained. In a steady state, the population difference implied by (168), (170) is

$$\rho_{aa} - \rho_{bb} = [(\Lambda_a/\gamma_a) - (\Lambda_b/\gamma_b)] \times [1 + (2\gamma_{ab}R/(\gamma_a\gamma_b))]^{-1}, \quad (171)$$

which shows clearly the effect of o.f. saturation.

Since the rate constant (170) depends on position z through normal modes $U_n(z)$, the population difference (171) also depends on position, and may be said to have "holes" burned in it. Consequences of this for mode competition can be discussed along the lines followed in Sec. 10. Under most conditions the behavior will correspond to the weak-coupling case, although if the excitation density were such that $-N_2$ were larger than \bar{N} the strong coupling case might be realized.

Combining Eqs. (165), (167), (171), and (41) we obtain a polarization

$$P(z, t) = \varphi(\rho_{ab} + \rho_{ab}^*) \quad (172)$$

and the coefficients S_n and C_n which enter the amplitude and frequency determining Eqs. (13) and (14) are

$$S_n = -\frac{2}{L} (\varphi^2/\hbar) \gamma_{ab} \mathcal{L}(\omega - \nu_n) E_n \times \int_0^L dz [U_n(z)]^2 N(z, t) [1 + (2\gamma_{ab}R(z)/\gamma_a\gamma_b)]^{-1} \quad (173)$$

and

$$C_n = [(\omega - \nu_n)/\gamma_{ab}] S_n. \quad (174)$$

These expressions depend nonlinearly on the mode amplitudes E_n because of saturation. However, since we have already neglected the beat frequency pulsations of $\rho_{aa} - \rho_{bb}$ which lead to terms with Δ in the denominator, it might not be consistent to keep any terms in R which are off-resonance by more than about $|\omega - \nu_{\mu}| \sim (\gamma_{ab}\Delta)^{1/2}$.

We will now consider only single-frequency operation, for which our Eqs. (173)–(174) are essentially exact. Then a single summand $\mu = n$ contributes to R and we find

$$S_n = -\frac{2}{L} \varphi^2 \hbar^{-1} \gamma_{ab} E_n \int_0^L dz [U_n(z)]^2 N(z) [\gamma_{ab}^2 + (\omega - \nu_n)^2 + \gamma_{ab}^2 (\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b)]^{-1}. \quad (175)$$

The integral may be easily calculated if $N(z)$ is a slowly

varying function of position, and we find

$$S_n = -\varphi^2 \hbar^{-1} \gamma_{ab} \bar{N} \mathcal{L}(\omega - \nu_n) f(w) E_n, \quad (176)$$

where

$$w = [(\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b)] \gamma_{ab}^2 \mathcal{L}(\omega - \nu_n) \quad (177)$$

and

$$f(w) = (2/w) [1 - (1+w)^{-1/2}]. \quad (178)$$

If Eq. (175) for S_n is expanded to third order in E_n , we obtain a result in agreement with that given by Eqs. (56), (72) for the single-frequency case with no atomic motion. It should be noted that whether (175) is expanded or not, the amplitude of oscillation has a maximum for resonant tuning and falls off monotonically with detuning. There is no indication here that the double maximum of Sec. 8 might be a spurious one which arises from the neglect of fifth or higher order terms.

17. DISCUSSION OF DOPPLER BROADENING

The effect of atomic motion upon our equations is rather curious and warrants discussion which, for simplicity, will be given for single-frequency operation. As we have seen in Sec. 16, the optical properties of the medium may be described by a nonlinear susceptibility

$$\chi(\omega - \nu_n, E_n) = P_n / (\epsilon_0 E_n). \quad (179)$$

It would perhaps be plausible to hope that the following simple recipe would take atomic motion into account. Because an atom moving with velocity v sees a Doppler shifted frequency, in the laboratory frame of reference it effectively has its resonance frequency shifted by $\omega v/c$. The effective susceptibility ought then to be

$$\chi_{\text{eff}} = \int_{-\infty}^{\infty} dv W(v) \chi(\omega - \omega(v/c) - \nu_n, E_n), \quad (180)$$

which can easily be expressed in terms of the $Z(\xi)$ function which is discussed in Sec. 6. The effective damping constant becomes

$$\gamma_{ab} [1 + (\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b)]^{1/2}$$

instead of γ_{ab} , and when this is much smaller than the Doppler parameter Ku , the line shape should be Gaussian with a normal Doppler width.

The above prescription is incorrect except to the first order in E_n if standing waves rather than running waves are involved in any way. This follows from a study of Eq. (70) contributing to the third-order polarization which involves a threefold integration over times τ' , τ'' , and τ''' . For single-frequency oscillation, the integrand contains

$$U_n(z - v\tau') U_n(z - v\tau' - v\tau'') U_n(z - v\tau' - v\tau'' - v\tau''') \times \exp[-i(\omega - \nu_n)\tau'] \exp[\pm i(\omega - \nu_n)\tau''']. \quad (181)$$

Each of the U_n 's is a standing wave which can be expressed as the sum of two running waves like $\exp \pm i(Kz - v\tau')$, etc. The physical interpretation is that in order to contribute to $P_n^{(3)}$ at time t an atom

has to interact three times with the o.f. field: first at $t''' = t - \tau' - \tau'' - \tau'''$, then at $t'' = t - \tau' - \tau''$, and finally at $t' = t - \tau'$. At each of these interactions the atom has a "choice" of interacting with one or the other of the two running waves. The τ dependence of a typical term of the integrand of (71) is then

$$\exp -\gamma_{ab}(\tau' + \tau''') - \gamma_a \tau'' \exp i(\nu_n - \omega)(\tau' \pm \tau''') \pm iKv\tau' \\ \pm iKv(\tau' + \tau'') \pm iKv(\tau' + \tau'' + \tau''').$$

The physical consequence of the appearance of terms involving $\pm Kv$ in the exponent is that each interaction involves a Doppler shift. Since we are interested in the case of a large Doppler width Ku , such a shift can take a given interaction very far from resonance even if $\omega = \nu_n$. This is expressed by the destructive interference of contributions to the integral at various value of τ' , τ'' , and τ''' . The interference will be least when the accumulated Doppler phase angle

$$\pm Kv\tau' \pm Kv(\tau' + \tau'') \pm Kv(\tau' + \tau'' + \tau''')$$

is zero. In order to obtain a nonvanishing spatial Fourier projection (8) for $P_n^{(3)}$, the choices \pm can not be all alike. The six remaining possibilities for the phase angle are

$$\pm Kv[\tau' + (\tau' + \tau'') - (\tau' + \tau'' + \tau''')] = \pm Kv(\tau' - \tau'''), \\ \pm Kv[\tau' - (\tau' + \tau'') + (\tau' + \tau'' + \tau''')] = \pm Kv(\tau' + \tau'''),$$

and

$$\pm Kv[-\tau' + (\tau' + \tau'') + (\tau' + \tau'' + \tau''')] = \\ \pm Kv(\tau' + 2\tau'' + \tau''').$$

As seen in Sec. 7, only the first two possibilities are able to lead to vanishing interference, and then only when $\tau' = \tau'''$.

Physically, one may say that a dominant type of process involves three interactions: first, one with a right (left) running wave at t''' , then one with a left (right) running wave at t'' , and finally one with a left (right) running wave at t' , with the time intervals obeying $t - t' = t'' - t'''$ so that the accumulated Doppler phase angle

$$\pm [Kv(t - t') + Kv(t - t'') - Kv(t - t''')]]$$

cancels out at time t .

The above cancellation of Doppler interference would not occur if waves running in only one direction were present. The nonlinear terms are much less broadened and weakened by Doppler effect for a standing wave maser oscillator than would be inferred from a study of nonlinear propagation alone. The double peak in the power as a function of tuning met in Sec. 8 can occur only because β_n (saturation) is not as much Doppler broadened as α_n (linear gain profile).

18. HOLE BURNING

In his discussion of maser action Bennett³¹ has made use of the notion of "hole burning." Since it aids the physical understanding of the rather complicated equations, we will now show how this phenomenon is described in the present work. As Bennett's treatment does not bring in the population pulsations of Sec. 15, we will base our discussion on the simplified theory of Sec. 16 in which pulsations of population were neglected.

In Sec. 16 the atoms had zero velocity. It is possible to generalize the discussion for the case of atoms having a velocity distribution $W(v)$ at the cost of further fairly plausible assumptions which are no worse than approximations already made. We deal first with those atoms which have a definite velocity v and which were excited at z_0 at time t_0 . The perturbation experienced by such an atom is

$$V(t) = -(\varphi/\hbar) \sum_{\mu} E_{\mu} U_{\mu}(z_0 + v(t - t_0)) \cos(\nu_{\mu}t + \varphi_{\mu}) \quad (181)$$

so that instead of seeing fields at frequencies ν_{μ} the moving atom sees fields at twice as many frequencies, i.e., $\nu_{\mu} \pm Kv$. The rate concept approach of Sec. 16 can not be used since the atoms characterized by different values of z_0 and t_0 experience different perturbations, i.e., the *phases* are not the same for the various members of the ensemble of atoms arriving at z at time t . It is plausible, however, to estimate the effect of saturation on the population difference $\rho_{aa}(v, t) - \rho_{bb}(v, t)$ by using an equation like (171) with a perturbation $V(t)$ given by (181) but with the terms involving z_0 and t_0 omitted. The replacement for the velocity-dependent rate constant $R(v)$ is then plausibly

$$R(v) = \frac{1}{8} \gamma_{ab} (\varphi/\hbar)^2 \sum_{\mu} E_{\mu}^2 [\mathcal{L}(\nu_{\mu} - \omega + Kv) \\ + \mathcal{L}(\nu_{\mu} - \omega - Kv)]. \quad (182)$$

If $v=0$, this reduces to the space average of Eq. (170). The corresponding population difference for atoms having velocity v is then

$$\rho_{aa}(v, t) - \rho_{bb}(v, t) \\ = W(v) [(\Lambda_a/\gamma_a) - (\Lambda_b/\gamma_b)] \\ \times [1 + 2\gamma_{ab}R(v)/(\gamma_a\gamma_b)]^{-1}. \quad (183)$$

A plot of (183) against v would show the assumed velocity distribution with "holes" burned into it due to o.f. saturation effects. These holes would be appreciable whenever $R(v)$ became comparable to $\gamma_a\gamma_b/(\gamma_a + \gamma_b)$ which could occur near $Kv = \pm(\nu_{\mu} - \omega)$, so that *two* holes could be burned for each cavity mode in oscillation. At a resonance, where $\nu_n = \omega$, the two holes for the n th mode would merge and reinforce one another. The holes in the velocity distribution would have been

³¹ W. R. Bennett, Jr., Appl. Opt. Suppl. 1, 24-61 (1962), especially pp. 58-59. It should be noted that the holes are in first instance burned in the curve of population difference versus velocity, and only indirectly in a curve of gain versus frequency.

seen in Sec. 15 if the integration over velocities had not been carried out so soon.

With the above approximate expression for $\rho_{aa}(v) - \rho_{bb}(v)$ we may use Eqs. (25) to calculate a pseudo-first-order value of $\rho_{ab}(z, v, t)$ and $P(z, v, t)$ in the manner of Sec. 16. The result will be the same as before, but with a velocity dependent reduction factor

$$[1 + 2\gamma_{ab}R(v)/(\gamma_a\gamma_b)]^{-1}$$

to express the effect of saturation, where $R(v)$ is given by (182). There are similar reductions in the related functions $S_n(v)$ and $C_n(v)$. The coefficients S_n and C_n which enter the equations of self-consistency [(13), (14)] result from integration of these quantities over velocity. Thus

$$S_n = -\varphi^2 \hbar^{-1} \bar{N} \gamma_{ab} E_n \int_{-\infty}^{\infty} dv W(v) \mathcal{L}(\omega - \nu_n + Kv) \times [1 + 2\gamma_{ab}R(v)/(\gamma_a\gamma_b)]^{-1}. \quad (184)$$

For single-mode operation, the rate $R(v)$ in the denominator of (184) contains the two terms given by Eq. (182). If an expansion is made to first order in $R(v)$ the integration over v can be done easily for a Maxwellian velocity distribution in the limit of large Doppler width Ku , and one gets results equivalent to those of Sec. 8. The possible dip in output power as a function of cavity tuning in single-mode operation can be interpreted as a consequence of the merging of the two holes at $Kv = \pm(\nu_n - \omega) \rightarrow 0$.

For multimode operation a similar calculation can be made with the complete expression (183) for $R(v)$. This will give the dominant terms of the equations of Sec. 9 for a Maxwellian velocity distribution and large Doppler width. However, the frequency locking terms involving ψ will be missing.

For two-mode oscillation, (184) leads to especially strong mode competition attributable to hole burning when $Kv = \nu_2 - \omega = \omega - \nu_1$, i.e., the traveling wave along $+z$ for mode 2 and the traveling wave along $-z$ for mode 1 are both in approximate resonance with the atomic transition frequency for an atom having velocity v . This effect can be correlated with the peak in θ for $\omega = \nu_{12}$ mentioned in Sec. 10.

19. APPROXIMATE HIGHER ORDER THEORY FOR SINGLE MODE OPERATION

It would be possible, but quite tedious, to extend the calculations of the text to fifth and higher order for the single-frequency case. The simpler approximate procedure outlined below may serve in the absence of such calculations. It was mentioned above that an expansion of (184) to first order in $R(v)$ reproduces the equations of single-mode operation correct to third order in E_n . If this expansion is not made one may hope to have equations which are valid for stronger signals. The v integration is complicated, and we will content ourselves here with two special cases (a) $Ku \gg |\nu_n - \omega| \gg \gamma_{ab}$

and (b) $\nu_n = \omega, Ku \gg \gamma_{ab}$. In the former case, one finds approximately

$$S_n = -\pi^{1/2} \varphi^2 \bar{N} E_n / (\hbar Ku) [1 + \frac{1}{4} (\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b)]^{1/2}, \quad (185)$$

while at resonance, when $\nu_n = \omega$,

$$S_n = -\pi^{1/2} \varphi^2 \bar{N} E_n / (\hbar Ku) [1 + \frac{1}{2} (\varphi E_n)^2 / (\hbar^2 \gamma_a \gamma_b)]^{1/2}, \quad (186)$$

and the merging of the two holes shows up in a simple manner through the doubling of the term expressing the effects of saturation. It will be remembered that a similar doubling of the coefficient β_n took place in Sec. 8 and was responsible for the possible dip in maser output versus cavity tuning. Although the more general behavior of output versus tuning implied by Eqs. (185)–(186) should be qualitatively correct, it must be remembered that rather uncontrolled approximations have been made in their derivation.

20. EXCITATION OF LOWER MASER LEVEL BY SPONTANEOUS DECAY OF UPPER MASER STATE

It was mentioned in Sec. 6 that the lower maser level could, at least in part, be excited by spontaneous emission from the upper. For the present this complication will be treated only in an approximation in which the rate concept is valid. For simplicity we ignore atomic motion, although the work of Secs. 16 and 19 suggests how this could be allowed for approximately. We write rate equations like (168)

$$\begin{aligned} \dot{\rho}_{aa} &= -\gamma_a \rho_{aa} + R(\rho_{bb} - \rho_{aa}) + \Lambda_a, \\ \dot{\rho}_{bb} &= -\gamma_b \rho_{bb} + R(\rho_{aa} - \rho_{bb}) + \Lambda_b + f \gamma_a \rho_{aa}, \end{aligned} \quad (187)$$

where the extra source term $f \gamma_a \rho_{aa}$ describes the effects of radiative cascade excitation of b from a assuming that a fraction f of the decays from a are to b . The Λ 's describe the uncorrelated excitation of the two levels. In a steady state one finds a population density difference

$$\rho_{aa} - \rho_{bb} = [(\Lambda_a/\gamma_a)(1 - f(\gamma_a/\gamma_b)) - (\Lambda_b/\gamma_b)] \times [1 + R\{\gamma_a(1 - f) + \gamma_b\}/(\gamma_a\gamma_b)]^{-1}, \quad (188)$$

which should be compared to Eq. (171). It will be seen that the effect of a nonvanishing branching ratio f is merely to change the unsaturated population difference (obtained for $R=0$), and also to modify the value of R for which a given degree of saturation would be obtained. The saturation parameter of Sec. 8 will be modified in an obvious fashion. Thus if $f=0$, a value of rate $R = \frac{1}{2} \gamma_a \gamma_b / \gamma_{ab}$ would cause 50% saturation, while if $f=1$ the value would be $R = \gamma_a$. It should be recalled that the dominant part of the third-order terms $S_n^{(3)}$ and $C_n^{(3)}$ are direct manifestations of saturation phenomena. At the present state of maser art, the decay constants γ_a and γ_b are not well enough known

for the effect of a nonvanishing value of f to be easily seen.

When population pulsations are taking place there will be a correlated time-dependent excitation of the lower level by cascade. It is possible that more interesting consequences than those obtained would result, and it is hoped to explore this possibility in a later paper.

21. OTHER SOURCES OF BROADENING

For some kinds of line broadening, especially in certain solid-state optical masers, one could adopt the recipe proposed in Sec. 17, and rejected for the case of Doppler broadening. If the effect of environment could be described by a distribution function for the atomic

resonance frequencies ω , an averaged nonlinear susceptibility could be used. This could also be done for the case of isotopic mixtures of the active atoms in gaseous masers.

Although γ_a and γ_b were introduced into our equations to describe spontaneous radiative decay of the states a and b , it is plausible that such phenomenological decay constants might also describe certain kinds of collision broadening. In that case, the γ 's would be functions of the pressure.³² A more detailed discussion of collision broadening for a gaseous optical maser will be given in another paper.

³² Evidence for such a dependence has recently been obtained by Javan and Szöke, Ref. 16.

Correlation Effects in Many Fermion Systems. II. Linked Clusters*

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(Received 31 January 1964)

In a previous paper a set of coupled equations was derived for the ground-state wave function and energy of a finite system of interacting Fermions. The equations are now modified so as to be more applicable to systems in which the number of particles becomes large. The resulting equations are shown to be equivalent to those obtained from many-body perturbation theory.

I. LINKED CLUSTERS

IN a previous paper,¹ a set of coupled equations was derived for the ground-state wave function and energy of a finite system of interacting Fermions. The wave function was expanded in terms of multiple-particle excitations on an uncorrelated zero-order state. The total energy E of the system appeared in the resulting equations and it was pointed out that this restricts the application of these equations to finite systems; in general, the restriction is to systems of small N . In the equations, the amplitudes for one-particle excitations are coupled to those for two-particle and three-particle excitations. The two-particle amplitudes are coupled to those for one-particle, three-particle, and four-particle excitations, and similarly for higher particle excitations. It was mentioned in I that it might be reasonable to approximate four-particle excitation terms, for example, as products of independent two-particle excitations.

It is shown here that four-particle terms involving two independently propagating pairs enter the equations in such a way as to eliminate the dependence of the two-particle excitation equations on the total

energy E , and similarly for the other excitations.² Explicit inclusion of products of independent excitations yields the equations of the linked cluster expansion. The resulting equations are shown to be the same as those obtained from many-body perturbation theory as formulated by Brueckner³ and by Goldstone.⁴

In I, the ground-state wave function is expanded as

$$|\psi\rangle = |\Phi_0\rangle + \sum_{\alpha,k} f(k;\alpha)\eta_k^+\eta_\alpha|\Phi_0\rangle + \sum_{\alpha,\beta,k,k'} f(kk';\alpha\beta)\eta_k^+\eta_{k'}^+\eta_\beta\eta_\alpha|\Phi_0\rangle + \dots \quad (1)$$

The unperturbed solution $|\Phi_0\rangle$ is a determinant composed of the N single-particle states which are lowest in energy.

Equations are then derived for $f(k;\alpha)$ and $f(kk';\alpha\beta)$ by inserting $|\psi\rangle$ from Eq. (1) into

$$H|\psi\rangle = E|\psi\rangle, \quad (2)$$

where H is written in the usual second-quantized form.¹

² I am indebted to Dr. A. M. Sessler for stressing the desirability of including products of independent pair excitations in the four-particle excitations, so as to make the resulting equations more applicable to systems of large N .

³ K. A. Brueckner, in *The Many Body Problem*, edited by C. DeWitt (John Wiley & Sons, Inc., New York, 1958).

⁴ J. Goldstone, Proc. Roy. Soc. (London) **A239**, 267 (1957).

* Work supported in part by the U. S. Atomic Energy Commission.

¹ H. P. Kelly and A. M. Sessler, Phys. Rev. **132**, 2091 (1963), hereafter referred to as I.