

Theory of a Three-Level Maser*

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In a three-level maser, a transition between two energy levels is saturated in order to produce an induced emission of power at a lower frequency corresponding to a transition between an intermediate energy level and one or the other of the two saturated levels. In this paper, certain effects are discussed which cannot be predicted from a theory in which the population differences alone are considered in this process. For instance, it is shown that, in some cases it is possible to obtain for the same system an induced emission of power at two frequencies given by the resonances between the intermediate energy level and both of the saturated levels. Also, it is shown that, even in the absence of inhomogeneous broadening of the spectral line, one can obtain a net induced emission at some portions of a resonant line and a net absorption

at other frequencies within the line widths. Such effects become important even in early stages of saturation in cases where $T_1 = T_2$ which is true for ordinary gaseous systems and individual spin systems in a majority of very dilute paramagnetic solids. A complete theory is discussed for a gaseous system and extended to two limiting cases of paramagnetic materials with $T_1 = T_2$ and $T_2 \ll T_1$ for each individual spin system. Furthermore, it is shown that for a saturating field of fixed frequency, the integral of induced power over the entire line is in full agreement with the results of a semiclassical treatment in which the population differences alone are considered. Some remarks are made as to the practicability of certain systems of this type.

INTRODUCTION

“MASER” action, the amplification of microwave power by means of stimulated emission of resonant radiation, was demonstrated some time ago for a beam of ammonia gas.¹ Another possibility for producing a similar effect is provided by a system where three energy levels are available such that a transition is allowed between the lowest and the highest of the three levels. If this transition is saturated by means of a large power, then a transition between the intermediate level and one or the other of the two saturated levels can be made to result in a net emission of radiation. When such emission is strong enough to overcome all losses of radiation, the system will become unstable and it can be used as an amplifier.

A three-level maser system of this type was discussed by Basov and Prokhorov,² who suggested its use in a molecular beam apparatus. Bloembergen has discussed the application of this technique to paramagnetic solids³ and such a system is reported to have been operated by Scovil, Feher, and Seidel.⁴

A semiclassical treatment of the mechanism of amplification of the microwave power can be given in terms of the averages of the population differences of the various levels in the presence of the saturating power. However, a detailed analysis of the subject reveals certain features which are not predictable from such a treatment. For instance, it is shown in Sec. III of the present paper that one could have a situation where even in the absence of inhomogeneous broadening of the spectral line, the induced power at the amplifying transition would appear in both emission and absorption

phases within different portions of the line widths. Such a result cannot be predicted on the basis of a change in the population differences alone. The presence of the saturating power will give rise to an admixture of the two saturated states such that in some cases it becomes inappropriate to distinguish the two states in order to speak in terms of the population differences involving these two levels.

The line shape at the amplifying frequency, and its dependence on the intensity and frequency of the saturating power as derived in this paper, at first sight may seem to contradict the results obtained from a treatment based on the population differences alone. However, it is shown that for the saturating power at a fixed frequency, the integral of the induced power over the entire line at the amplifying frequency, is in full agreement with the semiclassical calculations.

The purpose of the present paper is to present some aspects of the quantum-mechanical effects involved in a three-level maser. In order to reduce the problem to its essential points, first a complete analysis will be given, specifically for a gaseous system where the effects of the type mentioned above become of importance from early stages of saturation. Furthermore, the line structure and the thermal-relaxation processes, as applied to the three-level maser, are less involved in a gas than other systems such as paramagnetic solids. This aspect of the gaseous system makes it possible to give a systematic interpretation for various terms which unavoidably appear in a lengthy expression for the final results. Sections II through IV are devoted to this case. It is hoped that the detailed presentation of a gaseous system in this paper will give a better emphasis to the physical picture of the quantum-mechanical aspects of the problem. In Sec. V the results are extended to include two limiting cases for paramagnetic solids; namely, the cases with $T_1 = T_2$ and $T_2 \ll T_1$, for each individual spin system. In Sec. VI some remarks are

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¹ Gordon, Zeiger, and Townes, *Phys. Rev.* **99**, 1264 (1955).

² N. G. Basov and A. M. Prokhorov, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **28**, 249 (1955) [translation: *Soviet Phys. JETP* **1**, 184 (1955)].

³ N. Bloembergen, *Phys. Rev.* **104**, 324 (1956).

⁴ Scovil, Feher, and Seidel, *Phys. Rev.* **105**, 762 (1957).

made as to the extent of practicability of certain maser systems of this type.

II. SATURATION EFFECT IN A GAS

In this section, a method is described for calculation of the saturation effect in the resonant absorption of microwave power by a gas. Later in the paper, these calculations are extended to the case of the three-level system.

The molecular collisions will be assumed to be hard collisions, i.e., it will be assumed that the process of radiation is entirely interrupted upon each collision. A molecule which has undergone a collision will be found with an arbitrary phase in one of the stationary states of the isolated molecular energy states.⁵ Let us consider two stationary states, ψ_a and ψ_b , with ψ_b having a higher energy than ψ_a . Furthermore, assume that a transition is allowed between these two states and that a large radiation field is applied at a frequency near the resonance of this transition.

Immediately after a collision at a time t_0 , a molecule which is found in one of these two states will be affected by the presence of the radiation field and at a later time t , its wave function will take the form

$$\psi = a(t-t_0) \exp[-iE_a(t-t_0)/\hbar] \psi_a + b(t-t_0) \exp[-iE_b(t-t_0)/\hbar] \psi_b$$

and is a mixture of the two states. The equations of motion for a and b are:

$$\begin{aligned} \dot{a} &= yb e^{i(\omega' - \omega_0')t}, \\ \dot{b} &= -y^* a e^{-i(\omega' - \omega_0')t}, \end{aligned}$$

where $\hbar\omega_0' = E_b - E_a$, $y = \mu_{ab} E_0' / 2\hbar$, the radiation field having the form $E' = E_0' \sin \omega' t$ and μ_{ab} being the matrix element between the two states of the dipole moment in the direction of E_0' . It is assumed that no diagonal matrix elements exist for the dipole moment. In these equations the nonresonant terms of the form $e^{\pm i(\omega' + \omega_0')t}$ are ignored since they form rapidly varying perturbations and for $y \ll \omega_0'$ their effects are negligible.

If at the time t_0 the molecule is in the state $\psi = \psi_a$, the solution to the above equations becomes

$$\begin{aligned} \alpha &= \frac{1}{2} e^{\frac{1}{2}i(\omega' - \omega_0')(t-t_0)} \left[\left(1 - \frac{\omega' - \omega_0'}{2\gamma} \right) e^{i\gamma(t-t_0)} \right. \\ &\quad \left. + \left(1 + \frac{\omega' - \omega_0'}{2\gamma} \right) e^{-i\gamma(t-t_0)} \right], \quad (1a) \end{aligned}$$

⁵ Generally speaking, the wave function describing the state of each molecule is a mixture of the stationary states with arbitrary phase factors. The mixed wave function is such that the average, over statistical ensembles, of the probability of finding a molecule in a given stationary state is proportional to the Boltzmann distribution function. However, for the purposes of this paper, the random nature of the phase factors makes it possible to assume that the molecules exist in pure states with a Boltzmann population distribution.

$$b = \frac{iy^*}{2\gamma} e^{-\frac{1}{2}i(\omega' - \omega_0')(t-t_0)} [e^{i\gamma(t-t_0)} - e^{-i\gamma(t-t_0)}] \quad (1b)$$

where

$$\gamma = \frac{1}{2} [(\omega' - \omega_0')^2 + 4yy^*]^{\frac{1}{2}}.$$

The number of molecules which collide in a time interval dt_0 short compared with the mean collision time τ , but long compared with the duration of each collision, is Ndt_0/τ , where N is the total number of the molecules. A fraction, f , of these molecules, after collision, is found in a particular energy state, say E_a . This fraction is given by $f = g_a e^{-E_a/kT} / \sum g_i e^{-E_i/kT}$, where g_i is the degeneracy of the i th energy state and we assume a large number of energy states are available with energy separations in the microwave range. In other words, the total number of molecules which have undergone a collision in the time interval dt_0 and are found in a given energy state ψ_a is $n_a dt_0/\tau$, where n_a is the population of this state given by the Boltzmann distribution. The presence or absence of the strong radiation field has a negligible effect on the fraction f for gases considered in this paper. In particular, when the partition function is small, only a small fraction of the molecules exist in the states subject to the radiation field. Considering that the molecular collisions for the states having energy separations in the microwave region are diabatic in nature, a collision produces transition to any one of the many near-by energy states. Therefore, the presence of the radiation field cannot appreciably affect the distribution of the Ndt_0/τ molecules immediately after the collisions while only a small fraction of these molecules before collision have been disturbed by its presence. Even though, on the average, the population difference of the two states is affected by the applied field, immediately after collision a Boltzmann population distribution will be assumed for all states.

The assumption of random collisions with a mean collision time τ implies that the fraction of the molecules which have made a collision at a time t_0 and last for a time $t-t_0$ before making a second collision in a time interval dt is $(dt/\tau)e^{-(t-t_0)/\tau}$.

Considering that at the end of this time the probability of the exchange of a photon with the radiation field for those molecules which at the time t_0 are in either of the two states ψ_a or ψ_b is $|b(t-t_0)|^2$, where $b(t-t_0)$ is given by Eq. (1b), one obtains for the net absorbed power,

$$\begin{aligned} P &= (n_a - n_b) \frac{\hbar\nu}{\tau^2} \int_{-\infty}^t |b(t-t_0)|^2 e^{-(t-t_0)/\tau} dt_0 \\ &= (n_a - n_b) \frac{2\tau |y|^2 \hbar\nu}{1 + (\omega' - \omega_0')^2 \tau^2 + 4\tau^2 |y|^2}. \quad (2) \end{aligned}$$

Notice that in this treatment, the Boltzmann populations n_a and n_b appear in Eq. (2) because the popu-

lation distribution immediately after collisions is assumed to be a Boltzmann distribution and independent of the intensity of the applied radiation field. The saturation term in this equation arises from the fact that the transition probability is a periodic function of time, with a period which is a decreasing function of the strength of the applied field. Therefore, its average over the time distribution of collisions asymptotically approaches the value $\frac{1}{2}$ when the intensity of the resonating power is increased.

If one is interested in evaluating the average of the population difference between the two states subject to the radiation field, the above technique will yield

$$\begin{aligned} \bar{n}_a - \bar{n}_b &= (n_a - n_b) \frac{1}{\tau} \int_{-\infty}^t [1 - 2|b(t-t_0)|^2] \\ &\quad \times e^{-(t-t_0)/\tau} dt_0 \quad (3) \\ &= (n_a - n_b) \frac{(\omega' - \omega_0)^2 \tau^2 + 1}{1 + (\omega' - \omega_0)^2 \tau^2 + 4|y|^2 \tau^2}. \end{aligned}$$

The argument for the validity of this equation follows from noting that for the molecules $n_a dt_0/\tau$ which have made a collision at time t_0 and are found in the state ψ_a , the probability of their being found in the same state at the time t of a second collision is $[1 - |b(t-t_0)|^2]$. Also, within this time interval those molecules which were found in the state ψ_b immediately after collisions have a probability of $|b(t-t_0)|^2$ of being found in the state ψ_a . Furthermore, this procedure yields

$$\bar{n}_a + \bar{n}_b = n_a + n_b, \quad (4)$$

as expected.

These results are in agreement with derivations given previously⁶ for the power saturation by means of other treatments.

III. THE THREE-LEVEL SYSTEM

Let us suppose that a third energy level exists which lies somewhere in between the two levels subject to the saturating power. Let us change our notation slightly and designate the three states by ψ_1 , ψ_2 , and ψ_3 (Fig. 1). We assume that the saturating field is given by $E' = E_0' \sin \omega' t$, with ω' close to $\omega_0' = (E_3 - E_1)/\hbar$. Let us assume that a weak rf field $E = E_0 \sin \omega t$ is applied at a frequency ω close to $\omega_0 = (E_2 - E_1)/\hbar$.

To begin with, let us summarize some of the relevant results of the semiclassical treatment of the problem as mentioned in the Introduction.

In the absence of the saturating field and for the weak field at the frequency $\omega = \omega_0$, the power absorbed, from (2), is

$$P = (n_1 - n_2) 2h\nu |x|^2 \tau, \quad (5)$$

⁶ C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill Book Company, Inc., New York, 1955), Chap. 13. The parameter t in the Eqs. (13)-(74) through (13)-(79) of this reference should be replaced by $2t$.

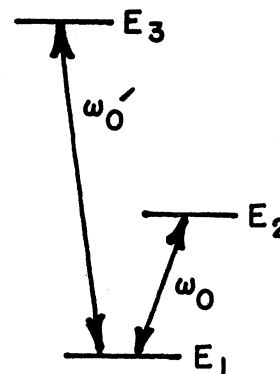


FIG. 1. States involved in the three-level system.

where $|x|^2 = |\mu_{12}|^2 E_0'^2 / (4\hbar^2)$, and it is assumed that $\tau^2 |x|^2 \ll 1$. When a large saturating power is present at a frequency ω' , the effect of this power on the induced transitions between ψ_1 and ψ_2 can be visualized roughly by noting that from (3) and (4), \bar{n}_1 decreases as the saturating power is increased. If, in Eq. (5), we substitute \bar{n}_1 for n_1 , since under our assumptions n_2 is not affected by the saturating field, it is then possible to obtain a situation where $\bar{n}_1 < n_2$, resulting in an induced emission of power. The expression for the emitted power at the frequency $\omega = \omega_0$ in terms of the saturating field intensity and for $\omega' = \omega_0'$, calculated as just outlined, is

$$P = (n_2 - n_3) h\nu |x|^2 \tau \left[\frac{4|y|^2 \tau^2 - p(2 + 4|y|^2 \tau^2)}{1 + 4|y|^2 \tau^2} \right], \quad (6)$$

where $|y|^2 = |\mu_{13}|^2 E'^2 / (4\hbar^2)$, and $p = (n_1 - n_2) / (n_2 - n_3)$. (Note that $|y|^2$ is proportional to the intensity of the saturating power.) This equation will be used later in comparing the limiting cases of the calculations which follow.

According to this equation, the threshold for the saturating power at which the absorption reduces to zero and the emission just begins is given by

$$2|y|^2 \tau^2 = p / (1 - p). \quad (7)$$

This relation indicates that an induced emission can be obtained at a resonance frequency corresponding to the energy separation of the states 1 and 2, if the parameter p is less than unity. Notice that for the cases where the energy separations are less than kT , one obtains $p \cong (E_2 - E_1) / (E_3 - E_2)$, which means that such emission can occur if the middle energy level is at most halfway between the energies of the states 1 and 3. Obviously, if $p > 1$, then an emission instead may be obtained at the resonance frequency of the states 2 and 3.

The optimum emitted power as a function of $|y|^2 \tau^2$ is obtained when this quantity becomes very large compared to unity, in which case (6) reduces to

$$P = 2[n_2 - \frac{1}{2}(n_1 + n_3)] h\nu |x|^2 \tau. \quad (8)$$

These results are obtainable, in exactly the same forms, from the treatment given by Bloembergen³ for

the paramagnetic solids, if one assumes that the rate of thermal relaxation is the same between various energy states.

In this analysis, however, the quantum-mechanical effects which influence the line shape at the amplifying frequency are ignored. In order to allow for such effects, we proceed as follows.

Let us consider a molecule which immediately after a collision is found in the state ψ_2 . After a certain time, as a result of the rf field at frequency ω , its quantum state becomes a mixture of the states ψ_1 and ψ_2 . On the other hand, such a mixed state is connected to the state ψ_3 via the radiation field at frequency ω' . Therefore a molecule which initially is found in the state ψ_2 has a finite transition probability to the state ψ_3 before its next collision. If at the time t_0 the state of the system is described by $\psi = \psi_2$, then at a later time t , ψ becomes

$$\psi = a_1(t-t_0) \exp\left[-\frac{iE_1}{\hbar}(t-t_0)\right] \psi_1 + a_2(t-t_0) \times \exp\left[-\frac{iE_2}{\hbar}(t-t_0)\right] \psi_2 + a_3(t-t_0) \exp\left[-\frac{iE_3}{\hbar}(t-t_0)\right] \psi_3.$$

Let us designate by $P_{ij}(t-t_0)$ the probability of a system which at the initial time t_0 is in the state ψ_i , and at a later time, t , makes a transition to the state ψ_j . Then $P_{23} = |a_3(t-t_0)|^2$ and $P_{21} = |a_1(t-t_0)|^2$, where a_3 and a_1 are the probability amplitudes as given in the above wave function subject to the initial conditions: $a_1(t-t_0) = a_3(t-t_0) = 0$ and $a_2(t-t_0) = 1$ for $t = t_0$. P_{21} is the probability of a direct transition between states ψ_2 and ψ_1 and is accompanied by emission of a photon at the frequency ω . On the other hand, P_{23} corresponds to a double photon exchange with the two radiation fields which induce the transition: one photon emitted at the frequency ω and one absorbed at the frequency ω' . Such a process is quite similar to the multiple-quantum transitions reported by various authors.⁷ The only difference is that in our case the intermediate energy state which serves to connect the initial and the final states has the lowest energy and the resonance condition is satisfied when the applied fields have their respective frequencies close to ω_0 and ω_0' . Thus, instead of absorption of two quanta [see reference 7], one photon is emitted whereas a second one is absorbed. The induced absorption and emission of radiation can be shown further by means of a study of the average of the dipole moment over the wave function, ψ , describing the mixed

states. From the expressions given below, one can evaluate the probability amplitudes a_1 , a_2 , and a_3 and show that the average dipole moment consists of two oscillating components at frequencies ω and ω' with such a phase relation to the applied fields that if it corresponds to an absorbing system at frequency ω' , it will correspond to an emitting system at frequency ω .

The reverse processes to the transitions defined by P_{23} and P_{21} arise from the system which immediately after collisions at the time t_0 are found in the state ψ_3 or ψ_1 and at a later time, t , make a transition to the state ψ_2 . It is intuitively obvious that the transition probabilities for the latter processes, P_{32} and P_{12} , satisfy the relations $P_{32}(t-t_0) = P_{23}(t-t_0)$ and $P_{21}(t-t_0) = P_{12}(t-t_0)$. This point is discussed further in the appendix of this paper.

In order to solve for the P_{ij} 's as defined above in terms of the applied field strengths and frequencies, let us consider the Schrödinger equation for the coefficients a_1 , a_2 , and a_3 :

$$\begin{aligned} \dot{a}_1 &= xa_2e^{i(\omega-\omega_0)t} + ya_3e^{i(\omega'-\omega_0')t}, \\ \dot{a}_2 &= -x^*a_1e^{-i(\omega-\omega_0)t}, \\ \dot{a}_3 &= -y^*a_1e^{-i(\omega'-\omega_0')t}, \end{aligned} \quad (9)$$

where $x = \mu_{12}E_0/(2\hbar)$ and $y = \mu_{13}E_0'/(2\hbar)$ as before.

In these equations the nonresonant terms, such as $e^{\pm i(\omega' \pm \omega_0)t}$, are ignored as they form rapidly varying perturbations and their effects are negligible.

A general solution to these equations can be obtained for arbitrary values of x and y . However, such a solution involves the roots of a polynomial of the third degree which cannot be put in a closed form if x is large and comparable to y . This solution is given in the appendix of the present paper. For small values of x , a solution correct to the first order in x can be calculated conveniently as described below, if the initial conditions are taken as those corresponding to P_{22} and P_{12} .

Let us consider that a molecule at the initial time t_0 is in the state $\psi = \psi_3$; then from (9) one obtains

$$a_2(t-t_0) = -x^* \int_{t_0}^t a_1 e^{-i(\omega-\omega_0)t'} dt'. \quad (10)$$

If $[(\omega-\omega_0) - (\omega'-\omega_0')]x$ is small compared to $|y|^2$, $a_1(t-t_0)$ can be solved by ignoring the terms in x in (9) and its solution is $a_1(t-t_0) = -b^*(t-t_0)$ where $b(t-t_0)$ is given in Eq. (1b). From this and the above equation, one obtains

$$\begin{aligned} P_{32}(t-t_0) &= |a_2(t-t_0)|^2 \\ &= \frac{|y|^2 |x|^2}{\gamma^2} \left[\frac{\sin^2[(\gamma-\Omega)(t-t_0)/2]}{(\gamma-\Omega)^2} + \frac{\sin^2[(\gamma+\Omega)(t-t_0)/2]}{(\gamma+\Omega)^2} \right. \\ &\quad \left. + \frac{\cos^2(\gamma(t-t_0)) + \frac{1}{2}\{\cos[(\gamma-\Omega)(t-t_0)] + \cos[(\gamma+\Omega)(t-t_0)]\}}{\gamma^2 - \Omega^2} \right], \end{aligned} \quad (11)$$

⁷ See, for instance, V. Hughes and L. Grabner, Phys. Rev. **79**, 829 (1950).

where $\Omega = \frac{1}{2}(\omega' - \omega_0) - (\omega - \omega_0)$, and $\gamma = \frac{1}{2}[(\omega' - \omega_0)^2 + 4|y|^2]^{\frac{1}{2}}$. Similarly, if one assumes that the molecule is initially in the state $\psi = \psi_1$, then a_1 will be the same as (1a) and from (10) one obtains

$$P_{12}(t-t_0) = |x|^2 \left[\frac{(1+a)^2 \sin^2[(\gamma-\Omega)(t-t_0)/2]}{(\gamma-\Omega)^2} + \frac{(1-a)^2 \sin^2[(\gamma+\Omega)(t-t_0)/2]}{(\gamma+\Omega)^2} - (1-a^2) \frac{\cos\gamma(t-t_0) - \frac{1}{2}\{\cos[(\gamma-\Omega)(t-t_0)] + \cos[(\gamma+\Omega)(t-t_0)]\}}{\gamma^2 - \Omega^2} \right], \quad (12)$$

where $a = (\omega' - \omega_0)/(2\gamma)$. The approximation used in the derivation of the above equations is entirely valid for the case under consideration in this paper, since at the conclusion x will be taken as arbitrarily small for an evaluation of the condition for a self-sustained oscillation at frequency ω , and we shall not be concerned with the saturation effect at the amplifying frequency. Also, it should be mentioned that these solutions happen to be correct for arbitrary values of x and y when $(\omega' - \omega_0) - (\omega - \omega_0) = 0$, and if γ is taken as

$$\frac{1}{2}[(\omega' - \omega_0)^2 + 4(|x|^2 + |y|^2)]^{\frac{1}{2}}.$$

This condition is satisfied in particular when both the applied fields have frequencies at their respective resonances.

From the above equations, the net induced absorption or emission of power at the frequency ω is calculated as follows:

First let us consider only the transition probabilities between states 2 and 1. According to the discussions presented in Sec. II, immediately after collisions, the population distribution is given by a Boltzmann distribution which prescribes a larger number of such systems in state 1 compared to systems in state 2. Since $P_{12}(t-t_0) = P_{21}(t-t_0)$, such transitions will always result in a net absorption of power at frequency ω given by

$$P_a = (n_1 - n_2) \frac{h\nu}{\tau^2} \int_{-\infty}^t P_{12}(t-t_0) e^{-(t-t_0)/\tau} dt_0. \quad (13)$$

The process which results is a net emission of power at this frequency, however, arises from transition probabilities P_{23} and P_{32} which correspond respectively to emission and absorption of photons of frequency ω in the double-quantum transitions as described above. According to the Boltzmann population distribution $n_2 > n_3$, and since $P_{23}(t-t_0) = P_{32}(t-t_0)$ the net emitted power due to this process is

$$P_e = (n_2 - n_3) \frac{h\nu}{\tau^2} \int_{-\infty}^t P_{32}(t-t_0) e^{-(t-t_0)/\tau} dt_0. \quad (14)$$

In other words, this is a process in which the transitions from a lower energy state, ψ_2 , to a higher energy state, ψ_3 , are accompanied by emission of the photons at frequency ω , whereas the reverse transitions from ψ_3 to ψ_2 take place by absorption of such photons. Ac-

cordingly, the Boltzmann distribution gives more weight to the emitted photons and the net result will be an emission.

The combined effect of these two types of transitions will yield, for the total emitted power,

$$P = P_e - P_a. \quad (15)$$

First let us consider a case where the state ψ_2 lies in energy very close to ψ_1 such that $n_1 \cong n_2$ and in the absence of the saturating power only a negligible absorption at the frequency ω takes place. Furthermore, assume that $(n_2 - n_3) \gg (n_1 - n_2)$, i.e., $\omega_0' \gg \omega_0$. In this case, P_a can be ignored in (15) since the presence of the saturating power will produce an emission proportional to $(n_2 - n_3)$ given by P_e and much larger than P_a . The quantity P_e can easily be evaluated by substituting P_{32} from (11) in Eq. (14) which, upon integration, yields

$$P_e = (n_2 - n_3) h\nu |x|^2 \tau \frac{|y|^2}{\gamma^2} \times \left\{ \frac{1}{2[1 + (\gamma - \Omega)^2 \tau^2]} + \frac{1}{2[1 + (\gamma + \Omega)^2 \tau^2]} + \frac{(\gamma^2 - \Omega^2) \tau^2 (2\gamma^2 \tau^2 + 1) - 1}{(1 + 4\gamma^2 \tau^2)[1 + (\gamma - \Omega)^2 \tau^2][1 + (\gamma + \Omega)^2 \tau^2]} \right\}. \quad (16)$$

When the intensity of the saturating power is of such magnitude that $|y|^2 \tau^2 \gg 1$, the third term in this equation becomes negligible and the emission line will appear as a doublet. The peak of each component of the doublet will occur when $\gamma - \Omega = 0$ or $\gamma + \Omega = 0$, which correspond to frequencies given by

$$\omega = \omega_0 + \frac{1}{2}(\omega' - \omega_0) \pm \gamma. \quad (17)$$

For $\omega' = \omega_0'$, (17) reduces to $\omega = \omega_0 \pm |y|$. The splitting of the emission line arises from the same effect as observed in the resonant modulation experiment in the molecule OCS.⁸ The presence of the saturating power gives rise to a modulation of the wave function between the two states 1 and 3 at an angular frequency γ [see Eq. (1b)] which is half the separation of the components of the doublet as obtained from (17).

The third term in Eq. (16) can be considered as due to the interference term originating from an overlap of

⁸ S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).

the components of the doublet. Its effect is important when the splitting of the doublet is not very large.

For a given value of the intensity of the saturating field, the emission line is largest when $\omega' = \omega_0'$. Under this condition, from (16), the behavior of the line shape as a function of $|y|^{2\tau^2}$ (which is proportional to the strength of the saturating power) is as follows:

Suppose that the quantity $|y|$ is increased continuously from zero by increasing the saturating power. For small $|y|^{2\tau^2}$, the emission shows only one maximum at the frequency $\omega = \omega_0$. The emission intensity at this frequency increases from zero as $|\lambda|^{2\tau^2}$ is increased, to an optimum value $P_e = \frac{2}{3}(n_2 - n_3)h\nu|x|^2\tau$ for $|y|^{2\tau^2} = \frac{1}{2}$. For $|y|^{2\tau^2} > \frac{1}{2}$, the peak intensity decreases while the emission line broadens. At $|y|^{2\tau^2} = 5/7$ the line begins to split and two maxima appear. For $|y|^{2\tau^2} > 5/7$, the separation of the two maxima increases and eventually, the emission will show as a doublet with the intensities at the peak of each component of the doublet approaching the value

$$P = \frac{1}{2}(n_2 - n_3)h\nu|x|^2\tau. \tag{18}$$

$$P_a = (n_1 - n_2)h\nu|x|^2\tau \left\{ (1+a)^2 \frac{1}{2[1+(\gamma-\Omega)^2\tau^2]} + (1-a)^2 \frac{1}{2[1+(\gamma+\Omega)^2\tau^2]} - (1-a^2) \frac{(\gamma^2 - \Omega^2)\tau^2(2\gamma^2\tau^2 + 1) - 1}{(1+4\gamma^2\tau^2)[1+(\gamma-\Omega)^2\tau^2][1+(\gamma+\Omega)^2\tau^2]} \right\}. \tag{19}$$

The net emitted power $P = P_e - P_a$ can then be calculated from Eqs. (19) and (16).

Notice that the same interference term appearing in the expression for P_e also shows as the third term of Eq. (19) for P_a .

For $|y|^{2\tau^2} \gg 1$, the absorption term, P_a , also presents two peaks, however, with an intensity ratio given by

$$\left(\frac{1+a}{1-a} \right)^2 = \left[\frac{[(\omega' - \omega_0')^2 + 4|y|^2]^{\frac{1}{2}} + (\omega' - \omega_0')}{[(\omega' - \omega_0')^2 + 4|y|^2]^{\frac{1}{2}} - (\omega' - \omega_0')} \right]^2,$$

which for $\omega' = \omega_0'$ is equal to unity and independent of $|y|$. The two peaks occur at the same frequencies as the centers of the emission doublet, P_e , given by (17).

An examination of (19) and (16) shows that in the limiting case where $|y|^{2\tau^2} \ll 1$, P_e approaches zero, while P_a approaches the usual absorption formula:

$$P = \frac{2(n_1 - n_2)h\nu|x|^2\tau}{1 + (\omega - \omega_0)^2\tau^2}.$$

Also, if $(\omega' - \omega_0')$ becomes large, i.e., the saturation power is tuned away from its resonance frequency, ω_0' , both components of the emission doublet P_e diminish in intensity while the peak of the stronger component of the absorption doublet P_a approaches the frequency ω_0 with a line shape approaching the absorption formula given above, and the other component of this doublet reduces to zero.

Therefore, the optimum emission occurs at $|y|^{2\tau^2} = \frac{1}{2}$ corresponding to an intensity of the saturating power, somewhat less than that required to split the emission line.

Although, in the semiclassical treatment, as mentioned at the beginning of this section, no allowance was made for the behavior of the line shape, it is interesting to compare the results with the peak intensities as calculated above. Notice that Eq. (6) for $n_1 \cong n_2$ and for $|y|^{2\tau^2} = \frac{1}{2}$ reduces to the same value as the optimum peak intensity calculated here. However, (6) predicts an optimum emission intensity when $|y|^{2\tau^2} \gg 1$, with a value twice as large as that obtained above at the peak of each component of the split doublet.

Let us now consider the case where ω_0 is not very small, i.e., $n_1 \neq n_2$ and $(n_1 - n_2)$ is comparable to $(n_2 - n_3)$. In this case, the absorption term, P_a , arising from transition probabilities P_{12} and P_{21} , will be superimposed on the emission term discussed above. If the expression (12) for P_{12} is substituted in (13), after integration one obtains:

Since the optimum intensity for the emission doublet given by (16) appears for $\omega' = \omega_0'$, let us consider, in what follows, this condition to be satisfied. In this case the parameter a , appearing in Eq. (19), is zero and for $|y|^{2\tau^2} \gg 1$, the two peak intensities for P_a become $(n_1 - n_2)h\nu(|x|^2\tau/2)$. Combining this result with (18), the total emitted power at the peak of each component of the doublet is therefore given by

$$P = P_e - P_a = [n_2 - \frac{1}{2}(n_1 + n_3)]h\nu|x|^2\tau. \tag{20}$$

This result should be compared with the Eq. (8) derived from the semiclassical treatment. They differ by a factor of two, which evidently originates as a result of the splitting of the emission line. However, such an agreement of the intensities is obtained under the condition where $|y|^{2\tau^2} \gg 1$, for which the separation of the two components of the doublet is large and the interference effect, as given by the third terms of Eqs. (16) and (19) is negligible. In order to study this effect, first let us examine the behavior of the induced power, P , at the frequency $\omega = \omega_0$ as a function of the intensity of the saturating power. As $|y|^{2\tau^2}$ is increased continuously from zero by increasing the strength of the saturating power, the emission term P_e reaches its maximum at $|y|^{2\tau^2} = \frac{1}{2}$. A further increase of power results in a decrease of P_e at this frequency, and eventually, for $|y|^{2\tau^2} \gg 1$, it approaches zero. However, the absorption term, P_a , at this frequency shows its maxi-

imum value for $|y|^2\tau^2 \ll 1$ and continuously decreases towards zero as $|y|^2\tau^2$ is increased. In fact, for $|y|^2\tau^2 = \frac{1}{2}$, where P_e is at its maximum, P_a is reduced by a factor of $\frac{1}{3}$ from its value at $|y|^2\tau^2 = 0$. In terms of the transition probabilities, the above behavior means that the average of the transition probability over the collision time from state ψ_2 to state ψ_1 , $\langle P_{21} \rangle$, decreases as the saturating power is increased, while $\langle P_{23} \rangle$ first increases from zero and has its maximum at $|y|^2\tau^2 = \frac{1}{2}$. For $|y|^2\tau^2 > \frac{1}{2}$, $\langle P_{23} \rangle$ becomes larger than $\langle P_{12} \rangle$. This fact immediately suggests an anomalous behavior for the necessary requirement of the population differences $(n_2 - n_3)$ and $(n_1 - n_2)$ for observation of the induced emission at the frequency ω_0 . From (16) and (19), for $\omega' = \omega_0'$ and $\omega = \omega_0$, one obtains

$$P = P_e - P_a$$

$$= 2(n_2 - n_3)\hbar\nu|x|^2\tau \frac{3|y|^2\tau^2 - p(1 + |y|^2\tau^2)}{(1 + 4|y|^2\tau^2)(1 + |y|^2\tau^2)}, \quad (21)$$

where the parameter p is

$$p = (n_1 - n_2)/(n_2 - n_3) \cong (E_2 - E_1)/(E_3 - E_2),$$

as before. This equation should be compared with the relation (6) of the semiclassical calculation.

For $|y|^2\tau^2(3 - p) > p$, Eq. (21) becomes positive, resulting in a net emission of power. This condition can be satisfied if p is less than 3, or

$$(E_2 - E_1) \leq 3(E_3 - E_2). \quad (22)$$

As long as the middle energy level is at most $\frac{3}{4}$ of the way up in between the lowest and the highest levels, then, in principle, amplification at frequency ω_0 may be possible. The intensity of the saturating power which yields an optimum emission at the frequency $\omega = \omega_0$ can be evaluated from the value of $|y|^2\tau^2$ for which Eq. (21) is maximized. For larger values of $|y|^2\tau^2$, Eq. (21) decreases monotonically towards zero. Accordingly, for $|y|^2\tau^2 \gg 1$, one always obtains an induced emission, however small, at the frequency $\omega = \omega_0$ if $p < 3$. On the other hand, Eq. (20), which holds for $|y|^2\tau^2 \gg 1$, shows that at the peak of each component of the doublet occurring at frequencies given by (17), emission results only if $p < 1$. Therefore, it is clear that for $1 \leq p \leq 3$, the induced power around ω_0 appears in both the absorption and emission phases, depending on the frequency of the applied field and its closeness to the resonance frequency ω_0 . A closer examination of the expression for the induced power shows that a similar behavior could also be encountered for $p < 1$.

This behavior, which occurs in the absence of an inhomogeneous broadening of the spectral line, cannot be explained in terms of a physical picture where a change in the average population differences of the various levels is taken as the factor responsible for producing an induced emission of power. However, in terms of the transition probabilities, such an effect

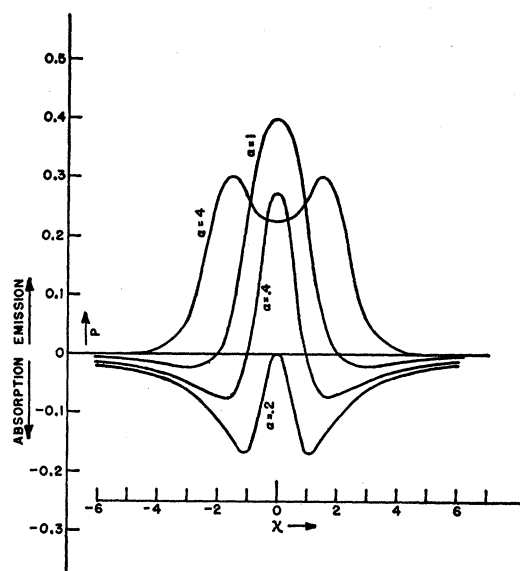


Fig. 2. The line shape at the amplifying frequency for various values of the strength of the saturating power. The ordinate represents an induced-emission coefficient defined by

$$P/[(n_2 - n_3)\hbar\nu|x|^2\tau]$$

and the abscissa is $\chi = (\omega - \omega_0)\tau$. The parameter α is proportional to the intensity of the saturating power and is defined by $\alpha = (|\mu_{13}|^2 E^2 / 4\hbar^2)\tau^2$. The frequency of the saturation power is taken at the resonance of the $1 \leftrightarrow 3$ transition. Also, $p = (n_1 - n_2)/(n_2 - n_3)$ is arbitrarily chosen as 0.5.

arises because at certain frequencies around ω_0 , $\langle P_{23} \rangle$ is larger than $p\langle P_{21} \rangle$ such that the result is an emission, whereas at some other frequencies the reverse is true and the result is an absorption.

In Fig. 2 a normalized line shape defined by $P/[(n_2 - n_3)\hbar\nu|x|^2\tau]$ is plotted against the dimensionless quantity $\chi = (\omega - \omega_0)\tau$ for $p = 0.5$ as a function of various values of $|y|^2\tau^2$.

In the treatments presented in this section, similar results would be obtained if ω_0 were defined as $(E_3 - E_2)/\hbar$ and a matrix element existed between states ψ_3 and ψ_2 . In this case, condition (22) should be replaced by $(E_3 - E_2) \leq 3(E_2 - E_1)$ for which an induced emission would be obtainable at a frequency near $\omega_0 = (E_3 - E_2)/\hbar$.

It is interesting to note that if transitions of the magnetic-dipole types are included, then it is possible to obtain simultaneously matrix elements connecting the state ψ_2 to both of the states ψ_1 and ψ_3 . In this case, if the separation of the middle energy level from the highest and the lowest levels is larger than $(E_1 - E_3)/4$, an induced emission near two frequencies $(E_2 - E_1)/\hbar$ and $(E_3 - E_2)/\hbar$ could be observed. This effect also cannot be accounted for on the basis of a change of the population differences alone.

In applying the expression for the induced power to the case where matrix elements exist between all the three levels, care has to be taken when p is very close to unity. In this case some of the nonresonant per-

turbations which were ignored in Eq. (9) as rapidly varying terms are no longer negligible and their effects should be included. A more general situation arises for any value of p when, in addition to the saturating power, two rf fields are present at frequencies close to $(E_2 - E_1)/\hbar$ and $(E_3 - E_2)/\hbar$. This case can be treated by evaluating the average dipole moment $\bar{\mu} = \int \psi^* \mu \psi d\tau$, where ψ is the wave function describing the state of a molecule at the time t subject to the initial condition prescribed by its state at the time t_0 immediately after a collision. The induced power can then be evaluated by calculating the susceptibility from an average of $\bar{\mu}$ over the time distribution of collisions and observing a Boltzmann population distribution immediately after collision.

IV. INTEGRATED LINE INTENSITY

The discrepancies with the calculations presented at the beginning of Sec. III evidently occur because of the presence of the saturating power giving rise to a drastic change in the line shape. However, a calculation on the basis of a change of the population difference should give a correct answer if, instead of intensity at a given frequency, the integrated line intensity is considered. This quantity is, in effect, an average intensity. One expects that a principle similar to that of "spectroscopic stability" is operative, such that any details of the line shape do not affect the integrated line intensity.

Equation (6) is calculated at the resonance frequency $\omega = \omega_0$. The result for the integrated line intensity will be the same if (6) is multiplied by a factor of π/τ . On the other hand, the integrated intensity for the actual line shape is $\int_0^\infty P d\omega$, where $P = P_e - P_a$ with P_e and P_a given by Eqs. (19) and (16). This integral can be evaluated easily by extending the integration from $-\infty$ to $+\infty$ and then closing the integral at infinity. The result is

$$\int_0^\infty P d\omega = \frac{\pi}{\tau} (n_2 - n_3) h\nu |x|^2 \tau \times \left\{ \frac{4|y|^2 \tau^2 - p[2 + 2(\omega' - \omega_0')^2 \tau^2 + 4|y|^2 \tau^2]}{1 + (\omega' - \omega_0')^2 \tau^2 + 4|y|^2 \tau^2} \right\}. \quad (23)$$

For $\omega' = \omega_0'$, this equation is the same as (6) when multiplied by π/τ . If we calculate (6) for the case where the frequency of the saturating power ω' , is taken off the resonance frequency ω_0' , the result becomes also the same as (23) in all details. Consequently, the semiclassical results, as derived from (6), are all correct for the integrated line intensity.

V. EXTENSION TO PARAMAGNETIC SOLIDS

The analysis as presented has been based on an assumption of hard molecular collisions and is complete for a gaseous system. Various effects of the types described are also present in any three-level maser

system such as in paramagnetic solids. However, the extent to which such effects may become of importance as a function of the intensity of the saturating power will depend on the type of thermal-relaxation mechanism operative in the material.

Two limiting cases in solids are encountered for which the above results can easily be extended; namely, when for each individual spin system $T_2 = T_1$ or $T_2 \ll T_1$. In solids or liquids with $T_1 = T_2$, the results obtained above will apply directly without any essential modification. It should be noted that the collision processes as described in Sec. II are equivalent, in other words, to assuming that when a molecule emerges from a collision it has essentially no memory of its past history. Such an assumption may be justified in all cases where the correlation time is very short, which implies that $T_1 = T_2$. These cases cover a majority of paramagnetic materials with very dilute concentration. The functional dependences, as expressed by Eqs. (15), (16), and (19) will apply in these cases to all the spin systems which have their center resonances at the frequency ω_0 . The presence of inhomogeneous broadening, which is usually the major source of the total width of the spectral line, can easily be allowed for by integrating (15) over a slowly varying function for the distribution of the local magnetic field.

Now let us consider a case where for each individual spin system $T_2 \ll T_1$. The exchange of energy between spin systems and the thermal bath takes place primarily as a result of spin-lattice interaction. This process is the major factor responsible for bringing about thermal equilibrium. The rate at which the system approaches equilibrium is given by $1/T_1$, and this approach may be a slow process if T_1 is long. On the other hand, the presence of spin-spin interaction will give rise to a simultaneous energy exchange between two spin systems without necessarily a net exchange of energy with the thermal bath. This process, in effect, shortens the lifetime of the states of each spin, and by including the adiabatic thermal processes which tend to disturb the phases of the spin system at random without causing a transition, the mean lifetime will be given by T_2 . We assume that $T_2 \ll T_1$. The principal feature of the treatment presented earlier lies in the presence of a transition of the type P_{23} . Such a transition becomes of importance if the strength of the saturating power is large enough to produce appreciable transition in a time shorter than T_2 where a spin system has a chance to show the effects which require its coherence. Let us first assume that the applied power is not large enough to produce such a transition. In this case, since the presence of the applied field does not result in an appreciable admixture of the states 1 and 3, it becomes permissible to speak in terms of the population differences of various states involving 1 and 3. Also, transitions between states 1 and 2 are of the ordinary type and the line shape at this power level is not affected by the presence of the saturating power.

If T_1 is long enough, the rate at which the energy absorbed from the saturating power is delivered to the lattice becomes slow and the population of the states 1 and 3 tends to become equalized. The semiclassical treatment, as discussed at the beginning of Sec. III, will apply rigorously and the situation is discussed by Bloembergen for an arbitrary rate of relaxation of the type T_1 between various energy states. For $|y|^2 T_2 T_1 \gg 1$, one obtains

$$(\bar{n}_2 - \bar{n}_3) = (\bar{n}_2 - \bar{n}_1). \quad (24)$$

Under the assumption of equal rates of relaxation between various levels this equation becomes equal to $[n_2 - \frac{1}{2}(n_1 + n_3)]$, where n_1 , n_2 , and n_3 are the populations of various states given by the Boltzmann distribution. At this power level, the induced-emission line shape for each individual spin system is the normal shape of the corresponding absorption line.

However, as soon as the applied power is increased further, such that saturation of levels 1 and 3 begins to set in a time T_2 , then transitions of the type P_{23} begin to become important at the expense of a change in P_{12} . Under this condition, in addition to power emitted as a result of a transition of the type P_{12} which is proportional to $(\bar{n}_2 - \bar{n}_1)$, an emission will occur because of P_{23} and proportional to $(\bar{n}_2 - \bar{n}_3)$. Considering that at this power level (24) holds, one obtains for the net power emitted:

$$P = (\bar{n}_2 - \bar{n}_3) h\nu [\langle P_{23} \rangle + \langle P_{21} \rangle],$$

where $\langle P_{23} \rangle$ and $\langle P_{21} \rangle$ are averages for the transition probabilities over the time distribution of the relaxation processes of the type T_2 . $\langle P_{23} \rangle$ and $\langle P_{21} \rangle$ will be given by $P_e / [(n_2 - n_3) h\nu]$ and $P_a / [(n_1 - n_2) h\nu]$ respectively, with P_e and P_a as given by Eqs. (16) and (19) and with τ substituted as T_2 . The result for $\omega' = \omega_0'$ becomes

$$P = (\bar{n}_2 - \bar{n}_3) h\nu |x|^2 T_2 \left\{ \frac{1}{1 + [|y| - (\omega - \omega_0)]^2 T_2^2} + \frac{1}{1 + [|y| + (\omega - \omega_0)]^2 T_2^2} \right\}. \quad (25)$$

For $\omega' \neq \omega_0'$ an extra term will appear in this expression because of the third term in P_a and P_e . Therefore, in the presence of a homogeneous broadening of the spectral line where for each individual spin system $T_2 \ll T_1$, for $|y|^2 T_2 T_1 \gg 1$, the power emitted is given by (25). This equation shows that for $|y|^2 T_2^2 \ll 1$ the emission has the usual shape of an absorption line; however, for $|y|^2 T_2^2 \gg 1$, the line appears as a doublet, resulting in a decrease of the induced emission at the frequency ω_0 . The presence of inhomogeneous broadening may easily be accounted for in this equation in the manner described for the case $T_2 = T_1$. Clogston has discussed the general case with arbitrary values of T_1 and T_2 , using the apparatus of density matrices. His

method can be shown to give results which are in agreement in all details for the two limiting cases as discussed in this paper.

From the above treatments we can conclude that in a three-level maser, there are various types of physical phenomena involved which cannot be explained on the basis of a change in the population differences alone. In particular, when $T_1 = T_2$ or in any case where a saturation is set up in the time T_2 , the states 1 and 3 will be so mixed that it becomes inappropriate to distinguish states 1 and 3 in order to speak only in terms of the population differences.

It should also be pointed out that, generally speaking, a three-level maser can be described as a type of monochromatically excited fluorescent effect. The two-quantum transitions of the type P_{23} involved in this process are similar to Raman-type scattering phenomena.

VI. CONCLUDING REMARKS

An obvious extension of a three-level maser can be made in systems where a fourth energy level is present with an energy larger than E_3 . In this case, the power induced at the amplifying frequency can be enhanced somewhat by applying a second saturating power at a resonant frequency close to $(E_4 - E_3)/\hbar$. For $T_1 = T_2$, the emission occurs as a result of the multiple-photon processes in the manner described for the three-level system. The expression for the emitted power is given in Appendix II for a special case where the frequencies of all the applied fields are taken at their respective resonances. A similar extension can also be made for systems with more than four energy levels where matrix elements exist between the required levels.

There exists a variety of substances with at least three energy levels where an induced emission of radiation could be observed on the basis of the mechanism described in this paper. However, the requirements for obtaining sufficient intensity to overcome other types of losses of the microwave power for the purpose of amplification are generally severe.

One is not limited only to electron-spin paramagnetism in solids for this type of amplification. For instance, direct nuclear quadrupole transitions in a crystal such as I_2 provide the required energy levels and at low temperature the condition for amplification may be satisfied. Such a crystal is not lossy and can be used with a large filling factor.

As far as gaseous states are concerned, for ordinary molecules, amplification may be obtained if the separation of levels 1 and 3 is large and in the millimeter range of frequency. The practicability of such systems for a CW operation will therefore require the availability of CW power in this range of frequency. For instance, the rotational transition $J=2 \leftarrow 1$ in the first excited state of the bending mode of vibration in

the molecule HCN could be used as the saturating transition. The direct transition between the l -type doublets with $\Delta J=0$ will then provide amplification at two frequencies corresponding to splittings of $J=1$ or $J=2$ levels. There are other molecules which can be used similarly. It should be noted that in these cases the expression for the emitted power is given essentially by Eq. (16), since P_a becomes negligible compared to P_e in (15) as soon as a reasonable saturating power is applied. Such systems cannot be used at very low temperatures which, at present, seem to be required for solids.

An interesting mechanism which provides the transitions of the required types in symmetrical-top molecules is encountered when a nucleus with large quadrupole coupling is present in the molecule. The hyperfine structure will give rise to forbidden transitions with $\Delta J=\pm 2$ or $\Delta J=\pm 3$ and can be used as the saturating transition. An allowed transition with $\Delta J=\pm 1$ can then be used as the active transition. For example, the molecule ICN seems to be a good example for this type of operation. Unfortunately, saturation of the $J=3\leftarrow 0$ transition in this molecule does not yield sufficient intensity for a convenient amplification at the $J=1\leftarrow 0$ transition. However, observation of the resonance line at the allowed transition and its dependence on the frequency of the saturating power seems to constitute a convenient way for detecting such types of forbidden transitions.

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APPENDIX I

A general solution of Eq. (9) can be calculated as follows.

Let us substitute in (9) $a_j = A_j e^{i(\lambda+\chi_j)t}$, where the a_j 's are the probability amplitudes with the subscript j standing for the levels 1, 2, and 3. If we then take $\chi_1=0$, $\chi_2=-(\omega-\omega_0)$, and $\chi_3=-(\omega'-\omega_0')$, Eq. (9) reduces to the following secular form:

$$\begin{aligned}\lambda A_1 + ix A_2 + iy A_3 &= 0, \\ -ix^* A_1 + (\lambda + \chi_2) A_2 &= 0, \\ -iy^* A_1 + (\lambda + \chi_3) A_3 &= 0.\end{aligned}$$

These equations can be solved simultaneously for the A_j 's if the determinant of their coefficients vanishes. This condition yields an equation of the third order in

λ , with real roots. With each root, one of the amplitudes A_j can be assigned arbitrarily. Accordingly, by writing $a_j = e^{ixt} \sum_k A_{jk} e^{i\lambda_k t}$, one obtains a general solution of (9) in terms of three unknown constants. The values of these constants can be chosen to satisfy the initial conditions.

Unfortunately, for arbitrary values of x and y , the roots of the secular equation cannot be expressed in a closed form except for $(\omega' - \omega_0') = (\omega - \omega_0)$. However, if x is small and $x \ll y$, a solution to the first order in x can be found easily and the relevant probability amplitudes in this order are given as follows: For the initial conditions that $a_3=1$, $a_2=a_1=0$ at $t=t_0$, one obtains

$$a_{32} = \frac{x^* y}{2\gamma} \left\{ \frac{e^{i(\gamma+\Omega)(t-t_0)} - 1}{\gamma + \Omega} + \frac{e^{-i(\gamma-\Omega)(t-t_0)} - 1}{\gamma - \Omega} \right\},$$

where the first subscript in a_{32} is used to indicate the initial condition, and γ and Ω are the same quantities as defined in Eq. (11). For $a_2=1$, $a_3=a_1=0$ at $t=t_0$, a_{23} can be evaluated in the same way and is found to be related to a_{32} by

$$a_{23} = \frac{xy^*}{yx^*} e^{-i[(\omega' - \omega_0') - (\omega - \omega_0)](t-t_0)} a_{32}.$$

Also, for initial conditions $a_1=1$, $a_2=a_3=0$ at $t=t_0$, one obtains

$$a_{12} = i \frac{x^*}{2\gamma} \left\{ \frac{(\gamma - \frac{1}{2}\chi_3)[e^{i(\gamma+\Omega)(t-t_0)} - 1]}{\gamma + \Omega} - \frac{(\gamma + \frac{1}{2}\chi_3)[e^{i(\gamma-\Omega)(t-t_0)} - 1]}{\gamma - \Omega} \right\}.$$

Finally the probability amplitude a_{21} defined by $a_2=1$, $a_1=a_3=0$ at $t=t_0$ is found to be related to a_{12} by

$$a_{21} = -\frac{x^*}{x} e^{-i(\omega - \omega_0)t} a_{12}.$$

Notice that from these probability amplitudes one obtains $P_{23}(t-t_0) = P_{32}(t-t_0)$ and $P_{21}(t-t_0) = P_{12}(t-t_0)$ as expected.

APPENDIX II

For a four-level maser, the probability amplitudes can be found with similar manipulations. Let us consider the case where all of the applied fields are at their respective resonances and the saturating powers are chosen such that $|\mu_{34}|E''/(2\hbar) = |\mu_{13}|E'/(2\hbar) = |y|$ and $|\mu_{12}|E/(2\hbar) = |x| \ll |y|$ with E , E' , and E'' as defined in Fig. 3. Under these conditions, the probability amplitudes can be evaluated to the first order in x ,

FIG. 3. States involved in a four-level system. The applied fields are: $\mathbf{E} = \mathbf{E} \sin \omega t$, with

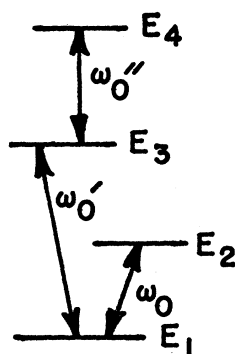
$$\omega \cong \omega_0 = (E_2 - E_1)/\hbar;$$

with

$$\omega' \cong \omega_0' = (E_3 - E_1)/\hbar;$$

with

$$\omega'' \cong \omega_0'' = (E_4 - E_3)/\hbar.$$



emitted at the frequency ω_0 :

$$P = |x|^2 \tau \hbar \nu \left\{ (n_2 - n_3) \frac{6|y|^2 \tau^2}{(1+8|y|^2 \tau^2)(1+2|y|^2 \tau^2)} + (n_2 - n_4) \left(\frac{1}{2(1+8|y|^2 \tau^2)} - \frac{1}{(1+2|y|^2 \tau^2)^2} + \frac{1}{2} \right) - (n_1 - n_2) \left(\frac{1}{2(1+8|y|^2 \tau^2)} + \frac{1}{(1+2|y|^2 \tau^2)^2} + \frac{1}{2} \right) \right\}.$$

and their averages over the time distribution of collisions yield the following expression for the power

This expression shows the characteristic behavior of the interference effect as pointed out in some detail in Sec. III of this paper.

Annihilation of Positrons in Gases*†

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The angular correlation of the two-photon annihilation of positrons annihilating in gases at high pressures demonstrates the two modes of decay—(1) from a bound state, positronium, and (2) from an unbound state—in all gases tested that have low-lying energy states. The increase in the narrow component in argon, plus suitable thermalizing gases, due to the application of a dc magnetic field obeys the theoretical increase due to the mixing of the 1^1S and 1^3S $m=0$ states. The increase in positronium formation due to the application of a dc electric field is observed. Preliminary evidence for the existence of $n=2$ states of positronium is seen.

I. INTRODUCTION

THE study of the process of positron annihilation by measurement of the angular correlation of the two quanta from the annihilation event has been used successfully in solids.¹⁻⁵ Prior to the present research, no precision experiment on the angular correlation of the two quanta from the annihilation events in gases has been reported. When positronium, the bound state of a positron and electron, is formed, the singlet state (1^1S) decays by two quanta and the triplet state (1^3S) decays by three quanta. The half-life for 1^1S positronium is

1.25×10^{-10} sec and for 1^3S is 1.4×10^{-7} sec.⁶ In this work the distribution in angle is usually limited to within ten milliradians of 180° with particular attention to the effect of a dc magnetic and/or a dc electric field on the events within two milliradians of 180° . The pressures used were from one to 28 atmospheres. The experiments on the magnetic quenching of orthopositronium done previously in various gases⁷⁻¹⁰ have been extended to higher fields. The effects on the angular correlation and magnetic field dependence of argon by adding thermalizing and/or quenching gases has been studied. Some experimental evidence for the $n=2$ states of positronium has been found. Some of the methods used here found immediate extensions, and those relating to parity nonconservation have already appeared in print.¹¹ However, for the sake of clarity, they are included here.

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¹ DeBenedetti, Cowan, Konnecker, and Primakoff, Phys. Rev. **77**, 205 (1950).

² Page, Heinberg, Wallace, and Trout, Phys. Rev. **98**, 206 (1955).

³ Lang, DeBenedetti, and Smoluchowski, Phys. Rev. **99**, 596 (1955).

⁴ A. T. Stewart, Phys. Rev. **99**, 594 (1955).

⁵ L. A. Page and M. Heinberg, Phys. Rev. **102**, 1545 (1956).

⁶ M. Deutsch, *Progress in Nuclear Physics* (Butterworths-Springer, London, 1953), Vol. 3, p. 131; S. DeBenedetti and H. C. Corben, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1954), Vol. 4, p. 191.

⁷ M. Deutsch and E. Dulit, Phys. Rev. **84**, 601 (1951).

⁸ T. A. Pond and R. H. Dicke, Phys. Rev. **85**, 489 (1952).

⁹ T. A. Pond, Phys. Rev. **85**, 489 (1952).

¹⁰ J. Wheatley and D. Halliday, Phys. Rev. **88**, 424 (1952).

¹¹ L. A. Page and M. Heinberg, Phys. Rev. **106**, 1220 (1957).