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Induced and Spontaneous Emission in a Coherent Field. IV

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In the previous articles of this series, dealing with the interaction between a number of molecules and the electromagnetic field in a resonant cavity, both the molecules and the field were treated by perturbation theory. The perturbation restriction on the field is removed in the present article, allowing large changes in the field, but the molecules are still assumed to undergo a small change during the time under consideration. The justification for this type of analysis, involving the generalization of the conventional concepts of induced and spontaneous emission, the applicability to a molecular amplifier during the buildup period, and the re-examination of a calculation by Serber and Townes concerning the fundamental limits of molecular amplification, is discussed.

Two different molecular distributions are considered. In one (the resonant case) all molecules have the same frequency as the cavity, and in the other (the nonresonant case) there is a uniform frequency distribution. The molecules are assumed to be initially in an emissive state. Several types of driving fields are considered. Expressions are obtained for the field operators by the solution of a Volterra integral equation, and expectation values are obtained for the field strength and field energy.

INTRODUCTION

IN the first three articles of the present series,¹ the interaction between a number of two-level atomic systems, referred to as molecules, and the electromagnetic field in a resonant cavity was analyzed by means of perturbation theory. The results were therefore correct only as long as both the molecules and the field underwent small changes in the course of time. In the present article we remove the perturbation-theory restriction on the field; that is, we analyze the problem in such a manner that the results are valid for large changes in the field. The perturbation restriction on the molecules is retained, however; they are assumed to undergo only slight changes during the time under consideration.

There is justification for undertaking an analysis of this type. From a practical viewpoint, the experimental situation in a molecular amplifier can be such that the

In the resonant case, both the coherent and incoherent fields increase exponentially after a sufficiently long time, no matter how small the initial gain is. Their ratio becomes constant and is equal to the number of photons in the driving field only in the absence of dissipation. An interesting related result is the fact that the signal-to-noise ratio for constant signal input power increases as the cavity dissipation increases. An estimate of the total time for which the theory is valid is obtained from a consideration of the energy emitted by the molecules. Contact is made with perturbation theory for sufficiently small gain and short time.

In the nonresonant case the effect of the molecules is shown to be that of a negative dissipation. In contrast to the resonant case, the gain becomes exponential only if the negative dissipation exceeds, in absolute value, the true dissipation. The ratio of induced to spontaneous emission is, in this case also, equal to the number of photons in the driving field only in the absence of dissipation. However, the signal-to-noise ratio for constant input power drops with increasing cavity dissipation.

molecules undergo only a small perturbation while the field is amplified manyfold; this is so when the initial signal is very weak, and the number of molecules is sufficiently large so that adequate output may be obtained with little emission per molecule. Also, there may be either a replacement of molecules before their characteristics are changed significantly or a restoration of the molecules to their initial state by some other method. It should be made clear, however, that the present article is not a complete analysis of a molecular amplifier. Thus, the amplifier achieves a steady-state condition after a buildup period; our theory does not apply to the steady-state conditions (which will be discussed in later work), but does apply to the buildup period when the only interaction taking place is that between a given group of molecules and the field.² From a theoretical viewpoint, it is interesting to note that the situation considered is the most general one in which a clear-cut differentiation may be made between induced

¹ I. R. Senitzky, *Phys. Rev.* **111**, 3 (1958); **115**, 227 (1959); **119**, 1807 (1960), hereafter referred to as I, II, and III, respectively.

² Restoration of the molecules to their initial states is not part of this interaction.

and spontaneous emission. As soon as higher order perturbation effects significantly alter the state of the molecules, it is no longer possible to separate induced and spontaneous emission, since one affects the other through the reaction on the molecules.³ The conventional textbook definition of induced and spontaneous emission refers to the lowest order effects with respect to both the molecules and the field. In the case, however, where the space into which the molecules radiate is enclosed by conducting walls and the field can build up, it is useful to introduce a more general definition, which will present itself in the course of the analysis. There is a further reason that makes the present discussion interesting from a theoretical viewpoint. The situation being considered (small molecular change but possibly large field change) is the same as that considered by Serber and Townes⁴ in a treatment of fundamental limits of molecular amplifiers, except for the fact that they do not consider cavity losses. It turns out that when losses are taken into account, the results obtained are different from theirs.

The notation and some of the preliminary aspects of the present article are the same as those of the earlier articles, but will be summarized briefly for the sake of completeness and intelligibility. There will be, however, a treatment of cavity dissipation which is formally different from that of the preceding two articles of the present series. In these, a special model was used for the loss mechanism. Since they were written, a method for treating dissipation more generally has been developed⁵; although the results obtained with both methods are identical, the general method has more appeal and will be used in the present analysis.

In the previous articles of the series, the cases considered were those in which all the molecules were in perfect resonance with the cavity (I and II) and also those in which there was a prescribed finite spread in the molecular frequencies (III).¹ In the present article we will consider two extreme cases: one in which all the molecules have the same frequency as the cavity, and another in which the frequency distribution is flat in the neighborhood of the cavity frequency. In other words, we will consider the perfectly resonant and completely nonresonant cases. As may be anticipated, the former will turn out to be the more interesting one. Part I consists of the preliminary development of the problem up to the point where a frequency distribution for the molecules is selected. Part II considers the resonant case, and Part III is devoted to the nonresonant case.

PART I

The electromagnetic field in the cavity of the single mode under consideration,⁶ with angular frequency ω ,

is described in the usual manner by

$$\mathbf{E} = -4\pi c \mathbf{u}(\mathbf{r})P(t), \quad \mathbf{H} = \nabla \times \mathbf{u}(\mathbf{r})Q(t), \quad (1)$$

where $\mathbf{u}(\mathbf{r})$ is a normalized function describing the spatial dependence of the field in the cavity, and $Q(t)$ and $P(t)$ are the quantum-mechanical field operators satisfying the commutator relationship $[Q(t), P(t)] = i\hbar$. Each molecule is considered to be a two-energy-level quantum-mechanical system coupled to the field through an electric dipole moment γ . The energy difference between the two levels of the m th molecule is $\hbar\omega_m$. The representation to be used is one in which the energy of the free molecules is diagonal, and the analysis will be performed in the Heisenberg picture.

The equations of motion may be derived from the Hamiltonian

$$H = H_f + H_l + H_{fl} + \sum_m H_m + 4\pi c P(\sum_m u_m \gamma_m + u_D D), \quad (2)$$

where H_f is the Hamiltonian of the electromagnetic field, H_l is the Hamiltonian of the loss mechanism, H_{fl} is the coupling term of field to loss mechanism, H_m is the Hamiltonian of the m th molecule, and the last term describes the coupling between molecules and field and between driving mechanism and field. The driving mechanism is represented by the classically prescribed dipole moment,

$$D(t) = d(t) \sin(\omega t + \theta), \quad (3)$$

where $d(t)$ varies much more slowly than $\sin(\omega t + \theta)$, but is left arbitrary at present for reasons which will be apparent later. The component of γ_m along \mathbf{u}_m is denoted by γ_m . Only the field Hamiltonian need be specified explicitly:

$$H_f = 2\pi c^2 P^2 + (\omega^2/8c^2)Q^2. \quad (4)$$

It is shown in reference 5 that the equation of motion for P is

$$\ddot{P} + \beta \dot{P} + \omega^2 P = F(t) - (\omega^2/c)(\sum_m u_m \gamma_m + u_D D), \quad (5)$$

be raised concerning the validity of a single-mode treatment. The justification for such a treatment depends, of course, on the magnitude of the losses that are being considered. The losses are greatest when the cavity walls absorb without reflection power radiated inside the cavity. Under those circumstances the cavity appears no different from free space to a molecule inside the cavity, if thermal radiation from the walls is ignored. The spontaneous transition probability in free space is $4\omega^3 \tilde{\gamma}^2/3\hbar c^3$, where $\tilde{\gamma}$ [defined by Eq. (12a)] is the absolute value of the molecular electric dipole moment. The expression for the spontaneous transition probability inside a cavity in which only a single mode is in resonance with—and coupled to—the molecule is [see Eq. (II 87)] $8\pi Q \tilde{\gamma}^2/\hbar V$, where Q and V are, respectively, the quality factor and volume of the cavity. The ratio of cavity to free-space spontaneous emission is therefore $3\lambda^3 Q/4\pi^2 V$. If we consider the situation in which $V \sim \lambda^3$, we see that free-space effects become comparable to single-mode cavity effects for $Q \sim 10$. For significantly larger values of Q , the single-mode treatment is justified. These larger values of Q are required by the restrictions explicitly imposed in the development of the theory in the present and preceding articles.

³ I. R. Senitzky, Phys. Rev. **121**, 171 (1961).

⁴ R. Serber and C. H. Townes, in *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960).

⁵ I. R. Senitzky, Phys. Rev. **119**, 670 (1960).

⁶ Since cavity losses are taken into account, a question might

where β is the cavity loss constant, related to the cavity quality factor Q_c by the relationship

$$\beta = \omega/Q_c; \quad (5a)$$

and where $F(t)$ is an operator expressing both the thermal and quantum-mechanical fluctuation properties of the loss mechanism. This operator is defined by

$$\langle F(f) \rangle = 0, \quad (6)$$

$$\langle F(t_1)F(t_2) \rangle = \frac{\hbar\omega^3\beta}{4\pi^2c^2} \left[i \frac{\mathcal{P}}{t_2 - t_1} + \pi\delta(t_1 - t_2)f(T) \right], \quad (7)$$

where \mathcal{P} indicates principle value, and

$$f(T) \equiv 1 + 2(e^{\hbar\omega/kT} - 1)^{-1}, \quad (8)$$

T being the temperature of the cavity. If P is known, Q may be obtained from Eqs. (2) and (4):

$$Q = -(4\pi c^2/\omega^2)\dot{P}. \quad (9)$$

The differential Eq. (5) may be recast into an integral equation more suitable for our purposes. Assuming $\beta/\omega \ll 1$, and ignoring the frequency shift of the cavity due to the losses (which is a second-order correction in β/ω), we have

$$P = p_D + p_F + p', \quad (10)$$

where

$$p_D = -\frac{\omega}{c} \mu_D \int_{-\infty}^t dt_1 D(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1), \quad (10a)$$

$$p_F = -\frac{1}{\omega} \int_{-\infty}^t dt_1 F(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1), \quad (10b)$$

$$p' = -\frac{\omega}{c} \sum_m \int_0^t dt_1 \gamma_m(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1). \quad (10c)$$

In transforming the differential equation to an integral equation, we have assumed that the loss and driving mechanisms have been coupled to the field from the time $t = -\infty$ and that the molecules have appeared in the cavity at time $t = 0$. It is worth noting that one could begin the analysis with Eq. (10), in which case no specific mechanism need be assumed but rather a prescribed driving field, given by p_D .

We have two more integral equations of motion that are essentially equivalent to the two differential equations of motion for γ_m and H_m obtainable from the Hamiltonian of Eq. (2):

$$H_m(t) = H_m^{[0]} + \frac{2\pi c \mu_m}{i\hbar} \int_0^t dt_1 \times \{ [H_m(t_1), \gamma_m(t_1)], P(t_1) \}, \quad (11)$$

$$\begin{aligned} \gamma_m(t) = & \gamma_m^{[0]}(t) + \frac{4\pi c \mu_m}{\hbar^2} \int_0^t dt_1 \int_0^{t_1} dt_2 \\ & \times U_m(t-t_1) [\gamma_m(t_1), [\gamma_m(t_2), H_m(t_2)] P(t_2)] \\ & \times U_m^{-1}(t-t_1), \end{aligned} \quad (12)$$

where

$$U_m(\tau) \equiv \exp[(i/\hbar)H_m^{[0]}\tau],$$

$\gamma_m^{[0]}$ is the dipole moment of the free molecule given by

$$\gamma_m^{[0]}(t) = \tilde{\gamma} \begin{pmatrix} 0 & e^{-i\omega_m t} \\ e^{i\omega_m t} & 0 \end{pmatrix}, \quad (12a)$$

$H_m^{[0]}$ is the Hamiltonian of the free molecule, and the notation $\{A, B\} \equiv AB + BA$ is used. Equations (9)–(12) describe completely the behavior of the system of molecules and field and may be taken as the starting point of our calculation.

We come now to the essential approximations that permit the solution of our problem. In view of our assumption that the molecules are affected only slightly by the interaction with the field, we replace the molecular variables γ_m and H_m by $\gamma_m^{[0]}$ and $H_m^{[0]}$, respectively, in the interaction term of Eq. (12). This is the first essential approximation. The second essential approximation consists of two steps: We ignore the commutator of $P(t_2)$ with both $\gamma_m^{[0]}(t_1)$ and $H_m^{[0]}(t-t_1)$, and bring it out either to the right or left of the molecular variables; then we replace the factor containing the molecular variables by its expectation value. The physical significance of the second approximation may be explained [after one notes that $\gamma_m(t)$ will be substituted into Eq. (10) and that our final results will be expectation values] by the statement that we are ignoring quantum-mechanical effects—or correlations of quantum-mechanical fluctuations—of higher order than the second. It should be pointed out that the compounding of second-order quantum-mechanical effects is not excluded by our approximation. These approximations are identical to the ones used in reference 5 in analyzing the interaction between a conservative system and a dissipation mechanism of large heat capacity in order to obtain a quantum-mechanical description of dissipation.

Carrying out the above approximations, we obtain in place of Eq. (12),

$$\begin{aligned} \gamma_m(t) = & \gamma_m^{[0]}(t) \\ & + \frac{8\pi c}{\hbar} \mu_m \tilde{\gamma}^2 \langle I_m \rangle \int_0^t dt_1 P(t_1) \sin\omega_m(t-t_1), \end{aligned} \quad (13)$$

where

$$I_m = \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}_m, \quad (13a)$$

the index indicating that this operator operates only on

the state vector of the m th molecule, and is the unit operator for the others. Substituting from Eq. (13) into Eq. (10), we have

$$P = p_D + p_F + p_\gamma + p, \quad (14)$$

where

$$p_\gamma = -\frac{\omega}{c} \sum_m u_m \int_0^t dt_1 \gamma_m^{[0]}(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin \omega(t-t_1), \quad (14a)$$

and

$$p = -\frac{8\pi\omega}{\hbar} \gamma^2 \sum_m u_m^2 \langle I_m \rangle \int_0^t dt_1 \int_0^{t_1} dt_2 \times P(t_2) e^{-\frac{1}{2}\beta(t-t_1)} \sin(t-t_1) \sin \omega_m(t_1-t_2). \quad (14b)$$

Equation (14) is an integral equation for only one unknown P . We cannot go further with it, however, until we choose a frequency distribution for the molecules.

PART II

We treat first the case in which there are N molecules all having the same frequency ω . The analysis of this case may be conveniently divided into three sections. In Sec. A we derive the operator expression for P , in Sec. B we study the expectation value of the field strength, and in Sec. C we analyze the expectation value of the energy.

Section A

Setting $\omega_m = \omega$ in Eq. (14b) and carrying out the t_1 integration first, we have

$$p = \frac{8\pi\omega}{\hbar\beta} u^2 \gamma^2 N \langle I \rangle \int_0^t dt_2 \times [1 - e^{-\frac{1}{2}\beta(t-t_1)}] P(t_2) \cos \omega(t-t_2), \quad (15)$$

where we have dropped an oscillatory term in the integrand (of the t_1 integral), and set

$$\sum u_m^2 \langle I_m \rangle = N u^2 \langle I \rangle. \quad (16)$$

We can now write Eq. (14) in the standard form of a Volterra integral equation:

$$P(t) = \psi(t) + \int_0^t dt_1 K(t-t_1) P(t_1), \quad (17)$$

where

$$\psi(t) = p_D + p_F + p_\gamma; \quad (17a)$$

and

$$K(t) = (2k^2/\beta)(1 - e^{-\frac{1}{2}\beta t}) \cos \omega t, \quad (17b)$$

with

$$k^2 = (4\pi/\hbar) \omega u^2 \gamma^2 N \langle I \rangle. \quad (17c)$$

We proceed to solve this integral equation. A well-known method of solution when the kernel is a function of $t-t_1$ consists of the utilization of the Laplace trans-

formation. Applying the Laplace transformation to both sides of Eq. (17), we have

$$\bar{P}(s) = \bar{\psi}(s) + \bar{K}(s) \bar{P}(s), \quad (18)$$

where

$$\bar{K}(s) = \mathcal{L}\{K(t)\} = \int_0^\infty dt e^{-st} K(t), \quad (19)$$

and similarly for the other functions. From Eq. (18) we obtain

$$\bar{P}(s) = \bar{\psi}(s) / [1 - \bar{K}(s)], \quad (20)$$

so that the solution of our integral equation is given by the inverse Laplace transformation of the right side of Eq. (20):

$$P(t) = \mathcal{L}^{-1}\{\bar{\psi}(s) / [1 - \bar{K}(s)]\} = \frac{1}{2\pi i} \int_{\epsilon-i\infty}^{\epsilon+i\infty} ds e^{st} \frac{\bar{\psi}(s)}{1 - \bar{K}(s)}, \quad (21)$$

where ϵ is a suitably chosen real positive number. (It should be pointed out that this method gives a solution for $t \geq 0$ only.) Equations (17b) and (19) yield

$$1 - \bar{K}(s) = \varphi(s) \{ (s^2 + \omega^2) [(s + \frac{1}{2}\beta)^2 + \omega^2] \}^{-1}, \quad (22)$$

where

$$\varphi(s) = (s^2 + \omega^2) [(s + \frac{1}{2}\beta)^2 + \omega^2] - k^2 [(s^2 - \omega^2) + \frac{1}{2}\beta s]. \quad (22a)$$

It is important, for further calculations, to find the roots of $\varphi(s)$. We can do this easily if we consider only those situations in which

$$k/\omega \ll 1. \quad (23)$$

Condition (23) is similar to the condition $\beta/\omega \ll 1$ which we have already assumed, the former applying to the gain and the latter applying to the loss. We do not want to make any comparison between k and β at this time; we will therefore treat both k/ω and β/ω as small quantities of the first order. It will be a simple matter to consider those cases where one is much smaller than the other in the final results. We therefore set a root of $\varphi(s)$, denoted by s' , equal to the sum of a zeroth- and first-order quantity (we have already ignored second-order quantities by neglecting the frequency shift due to the loss)

$$s' = s^{(0)} + s^{(1)}, \quad (24)$$

and obtain immediately

$$s^{(0)} = \pm i\omega, \quad (25)$$

$$s^{(1)} = -\frac{1}{4}\beta \pm (\beta^2 + 8k^2)^{\frac{1}{2}}.$$

This gives us the four roots of $\varphi(s)$:

$$s_1 = i\omega + \alpha_0, \quad s_2 = -i\omega + \alpha_0, \quad (26)$$

$$s_3 = i\omega + \alpha_1, \quad s_4 = -i\omega + \alpha_1,$$

where

$$\begin{aligned}\alpha_0 &= -\frac{1}{4}\beta + \frac{1}{4}(\beta^2 + 8k^2)^{\frac{1}{2}}, \\ \alpha_1 &= -\frac{1}{4}\beta - \frac{1}{4}(\beta^2 + 8k^2)^{\frac{1}{2}}.\end{aligned}\quad (26a)$$

The relationships

$$\alpha_0 + \alpha_1 = -\frac{1}{2}\beta \quad (26b)$$

and

$$\alpha_0\alpha_1 = -\frac{1}{2}k^2$$

will prove useful.

From Eqs. (21) and (17a) we see that P consists of a sum of three parts:

$$P(t) = P_D(t) + P_F(t) + P_\gamma(t), \quad (27)$$

where

$$P_A(t) = \mathcal{L}^{-1}\{\bar{p}_A(s)/[1 - \bar{K}(s)]\}, \quad (27a)$$

with A standing for either D , F , or γ . We derive the three parts separately. For the sake of simplicity, we consider first the case in which the driving field has a constant amplitude,

$$p_D'(t) = p_{D0} \cos(\omega t + \theta). \quad (28)$$

In terms of the driving mechanism, constant amplitude is achieved when the driving dipole moment is given by

$$D(t) = \frac{c\beta p_{D0}}{\omega u_D} \sin(\omega t + \theta) \equiv D_0 \sin(\omega t + \theta). \quad (29)$$

From Eq. (28) we obtain

$$\bar{p}_D'(s) = p_{D0} \frac{s \cos\theta - \omega \sin\theta}{s^2 + \omega^2}. \quad (30)$$

Substituting into (27a), we have

$$P_D(t) = \frac{1}{2\pi i} \int_{\epsilon - i\infty}^{\epsilon + i\infty} ds \frac{e^{st}(s \cos\theta - \omega \sin\theta)[(s + \frac{1}{2}\beta)^2 + \omega^2]}{\varphi(s)}. \quad (31)$$

The value of $P_D(t)$ is equal to the sum of the residues of the integrand at the four roots of $\varphi(s)$, which are given by Eq. (26). Neglecting quantities in the residues of higher order than the first we obtain, after some routine calculation

$$P_D'(t) = p_D'(t)(\alpha_0 e^{\alpha_1 t} - \alpha_1 e^{\alpha_0 t})/(\alpha_0 - \alpha_1). \quad (32)$$

The physical significance of this result will be discussed later.

We consider now the situation in which the driving mechanism has been acting from $t = -\infty$ to $t = 0$, and is then shut off suddenly when the molecules appear. We have, then,

$$p_D''(t) = p_D'(t)e^{-\frac{1}{2}\beta t}. \quad (33)$$

Using the same method as that with which Eq. (32) was derived, we have

$$P_D''(t) = p_D'(t)(\alpha_0 e^{\alpha_0 t} - \alpha_1 e^{\alpha_1 t})/(\alpha_0 - \alpha_1), \quad (34a)$$

$$= p_D''(t)(\alpha_0 e^{-\alpha_1 t} - \alpha_1 e^{-\alpha_0 t})/(\alpha_0 - \alpha_1). \quad (34b)$$

Finally, for future use, we will need the value of $P_D(t)$ when the driving field has an arbitrary attenuation

$$p_D'''(t) = p_D'(t)e^{-\frac{1}{2}at}, \quad (35)$$

but still with $a \ll \omega$. For this case we obtain, with the above method

$$P_D'''(t) = p_D'(t) \left[\frac{\alpha_0\alpha_1}{\alpha_0 - \alpha_1} \left(\frac{e^{\alpha_1 t}}{\alpha_1 + \frac{1}{2}a} - \frac{e^{\alpha_0 t}}{\alpha_0 + \frac{1}{2}a} \right) - \frac{a(\beta - a)e^{-\frac{1}{2}at}}{4(\alpha_0 + \frac{1}{2}a)(\alpha_1 + \frac{1}{2}a)} \right]. \quad (36)$$

It is easily seen that Eqs. (32) and (34) are special cases of Eq. (36).

We evaluate now the other two parts of $P(t)$ in Eq. (27), namely, $P_\gamma(t)$ and $P_F(t)$. Using Eq. (12a) for $\gamma^{[0]}(t)$ and substituting in (14a) we obtain, with previously used approximations,

$$p_\gamma(t) = \frac{\omega\tilde{\gamma}}{c\beta} (1 - e^{-\frac{1}{2}\beta t}) \sum_m u_m \begin{pmatrix} 0 & -ie^{-i\omega_m t} \\ ie^{i\omega_m t} & 0 \end{pmatrix}_m. \quad (37)$$

Since we are considering now the case in which $\omega_m = \omega$, the Laplace transform of this expression is given by

$$\bar{p}_\gamma(s) = \frac{\omega\tilde{\gamma}}{2c} \sum_m u_m \begin{pmatrix} 0 & -iA(\omega) \\ iA(-\omega) & 0 \end{pmatrix}_m, \quad (38)$$

where

$$A(\omega) \equiv [(s + i\omega)(s + \frac{1}{2}\beta + i\omega)]^{-1}.$$

Substituting in Eq. (27a) we obtain, by previous methods,

$$P_\gamma = \frac{\omega\tilde{\gamma}}{2c} \frac{e^{\alpha_0 t} - e^{\alpha_1 t}}{\alpha_0 - \alpha_1} \sum_m u_m \begin{pmatrix} 0 & -ie^{-i\omega t} \\ ie^{i\omega t} & 0 \end{pmatrix}_m. \quad (39)$$

For P_F we write simply

$$P_F(t) = \int ds \int_0^\infty dt_1 \int_{-\infty}^{t_1} dt_2 \mathcal{F}(t, s, t_1, t_2) F(t_2), \quad (40)$$

where

$$\mathcal{F}(t, s, t_1, t_2) \equiv \frac{\exp[s(t - t_1) - \frac{1}{2}\beta(t_1 - t_2)]}{\omega[1 - \bar{K}(s)]} \sin\omega(t_1 - t_2). \quad (40a)$$

We cannot (and have no need to) evaluate this expression any further, since we have no explicit expression for $F(t)$.

Section B

Now that we have the operator expression for P , we can calculate its expectation value. The expectation value of P_D is, of course, the same as P_D itself (since it is already a c number), and is given by either Eq. (32), (34), or (36), depending on the kind of driving field we are considering. In order to obtain the expectation

value of P_γ , we must describe the initial state of all the molecules. Setting the wave function for this state equal to a product of one-molecular wave functions, each being given by

$$\psi_m = a_{m1}\varphi_{m1} + a_{m2}\varphi_{m2} \quad (41)$$

in obvious notation, we obtain

$$\langle P_\gamma \rangle = -\frac{\omega}{c} \frac{e^{\alpha_0 t} - e^{\alpha_1 t}}{\alpha_0 - \alpha_1} \sum_m u_m |a_{m1} a_{m2}| \sin(\omega t + \theta_m), \quad (42)$$

where θ_m is the phase difference between a_{m2} and a_{m1} , and has its physical significance illustrated by the fact that

$$\langle \gamma_m^{[0]}(t) \rangle = 2 |a_{m1} a_{m2}| \bar{\gamma} \cos(\omega t + \theta_m). \quad (43)$$

The expectation value of P_F is obtained immediately from Eqs. (40) and (6):

$$\langle P_F \rangle = 0. \quad (44)$$

The final result for the expectation value of P is therefore given by

$$\langle P \rangle = P_D + \langle P_\gamma \rangle, \quad (45)$$

P_D standing for either P_D' , P_D'' , or P_D''' .

We now consider the physical significance of our results for the expectation value of P . It is clear that P_D is produced by the driving field, and $\langle P_\gamma \rangle$ is independent of the driving field. P_D is therefore (when multiplied by $-4\pi c$) the superposition of driving field strength and induced field strength, or the amplified driving field. Let us discuss it in greater detail.

Consider first the case of the constant driving field $p_D'(t)$, given by Eq. (28). Then the amplified field is $P_D'(t)$, given by Eq. (32). The amplifying factor is

$$(\alpha_0 e^{\alpha_1 t} - \alpha_1 e^{\alpha_0 t}) / (\alpha_0 - \alpha_1), \quad (46)$$

which, for sufficiently large time, becomes

$$-\alpha_1 (\alpha_0 - \alpha_1)^{-1} e^{\alpha_0 t}, \quad (47)$$

since α_0 is positive and α_1 is negative. We see that the gain is eventually exponential in time. (We must remember, of course, our original restriction on the time, which requires that it be less than that during which the molecules undergo substantial change.) If $k^2 = 0$, (i.e., no molecules in the cavity), then $\alpha_0 = 0$, $\alpha_1 = -\frac{1}{2}\beta$, and the complete amplification factor is identically unity, as it should be.

It is interesting to see under what conditions the amplification factor (46) reduces to a perturbation theory expression. For perturbation theory to be valid, the amplification factor must be only slightly larger than unity. This condition is satisfied when t is sufficiently small. However, if we want the time during which perturbation theory is valid to be longer than the cavity relaxation time β^{-1} , expression (47) must be only slightly larger than unity. Perturbation theory is therefore applicable (when the time under considera-

tion extends to values larger than the cavity relaxation time) if $\alpha_0 \ll \alpha_1$ and $t \ll \alpha_0^{-1}$. The former inequality implies that

$$k^2 \ll \beta^2, \quad (48)$$

so that

$$a_0 \approx k^2/\beta, \quad \alpha_1 \approx -\frac{1}{2}\beta[1 + 2(k^2/\beta^2)]. \quad (49)$$

The amplification factor (46) then becomes, up to the first power in both k^2/β^2 and $\alpha_0 t$,

$$1 + 2(k^2/\beta^2)[\frac{1}{2}\beta t - (1 - e^{-\frac{1}{2}\beta t})]. \quad (50)$$

Comparison with II¹ [Eq. (55)] shows complete agreement with the perturbation theory derivation used there.

We consider now the case in which the driving field is given by Eq. (33), i.e., it decays with the factor $e^{-\frac{1}{2}\beta t}$. This is the situation in which the driving mechanism is shut off at $t=0$. From Eq. (34a) the field becomes, for t sufficiently large,⁷

$$p_D'(t) (\alpha_0 e^{\alpha_1 t}) / (\alpha_0 - \alpha_1). \quad (51)$$

This result seems puzzling at first, for it indicates that no matter how small the number of molecules (which makes k^2 small), the gain due to the molecules seems to outweigh the decay due to the cavity losses, for sufficiently large t . The explanation lies in the very last phrase. Our analysis assumes that the molecules are affected only to lowest order by the field. The lowest-order induced emission of molecules that are all in resonance with the field increases with time. (The situation is different for a significant frequency spread, as shown in III.)¹ Therefore, in our analysis the amplifying property of the molecules increases indefinitely and eventually compensates for the cavity losses completely. The fewer the molecules, the longer it takes for this to happen. If k^2 is very small, the coefficient of the decaying exponential $e^{\alpha_1 t}$ is much larger than the coefficient of the increasing exponential $e^{\alpha_0 t}$ in expression (34a). One can see readily that this expression decreases at first. (Note that this is not so for expression (39) which refers to a constant driving field and is a monotonically increasing function of t .) It begins to increase after a time, the length of which depends on k^2 , and becomes large as k^2 becomes small. This time may be larger than the time during which our calculation is valid. An estimate of the time during which the analysis is valid will be given later in the discussion of energy. Beyond the time of validity, higher-order effects reduce the induced emission of the molecules, bringing about saturation of the amplification properties.

We consider finally, as far as the field is concerned, $\langle P_\gamma \rangle$ given by Eq. (42). It is the part of the field that is generated by the initial oscillation of the dipole moment

⁷ Note that the sufficiently large t in this case is larger than in the case of the constant driving field. Here t must satisfy the relationship $\alpha_0 e^{\alpha_0 t} \gg |\alpha_1| e^{\alpha_1 t}$, while for the case of the constant driving field we must have $|\alpha_1| e^{\alpha_0 t} \gg \alpha_0 e^{\alpha_1 t}$. Since $|\alpha_1| > \alpha_0$, the second inequality holds for smaller t .

of the molecules. If the molecules have all been prearranged to oscillate in phase, the $\langle P_\gamma \rangle$ may be large. If, however, the molecular phases are random, then $\langle P_\gamma \rangle$ vanishes.

Section C

We come now to a discussion of the expectation value of the energy in the field. We divide this section into three subsections. In subsection 1 we derive and discuss the expectation value of the energy; in subsection 2 we obtain an estimate of the length of time for which the present theory is valid; in subsection 3 we compare the coherent to the incoherent field.

Subsection 1

The expectation value of the field energy is given by

$$\langle H_f \rangle = 2\pi c^2 \langle P^2 \rangle + (\omega^2/8\pi c^2) \langle Q^2 \rangle. \quad (52)$$

It is easy to show that, when averaged over a cycle, the magnetic energy (the Q^2 term) is approximately equal to the electric energy (the P^2 term). We therefore need to calculate only $\langle P^2 \rangle$, average over a cycle, and multiply by $4\pi c^2$ in order to obtain the total energy.

For the square of the field, we have

$$\langle P^2 \rangle = \langle P_D^2 \rangle + \langle P_\gamma^2 \rangle + \langle P_F^2 \rangle, \quad (53)$$

the cross terms vanishing in the expectation value. The first term on the right side of Eq. (53) is obtained immediately from P_D , since the latter is a c number given by Eq. (32), (34), or (36), depending on the type of driving field being considered. It is clear that $4\pi c^2 \times \langle P_D^2 \rangle_{av}$ is the sum of the driving field energy and induced emission energy. (Note that the induced emission energy as now defined is not merely the lowest-order perturbation term.) For the three types of driving field we have the following relationships:

$$4\pi c^2 \langle P_D'^2 \rangle_{av} = E_D' [(\alpha_0 e^{\alpha_1 t} - \alpha_1 e^{\alpha_0 t}) / (\alpha_0 - \alpha_1)]^2 \quad (54)$$

for the case in which the driving field is of constant amplitude, where E_D' is the energy of the driving field (i.e., the field in the absence of the molecules);

$$4\pi c^2 \langle P_D''^2 \rangle_{av} = E_D' [(\alpha_0 e^{\alpha_0 t} - \alpha_1 e^{\alpha_1 t}) / (\alpha_0 - \alpha_1)]^2 \quad (55)$$

for the case in which the driving mechanism is turned off at $t=0$, and therefore the driving-field energy is given by

$$E_D'' = E_D' e^{-\beta t}; \quad (56)$$

and lastly

$$4\pi c^2 \langle P_D'''^2 \rangle_{av} = E_D' \left\{ \frac{\alpha_0 \alpha_1}{\alpha_0 - \alpha_1} \left[\frac{e^{\alpha_1 t}}{\alpha_1 + \frac{1}{2}a} - \frac{e^{\alpha_0 t}}{\alpha_0 + \frac{1}{2}a} \right] - \frac{a(\beta - \alpha)e^{-\frac{1}{2}at}}{4(\alpha_0 + \frac{1}{2}a)(\alpha_1 + \frac{1}{2}a)} \right\}^2 \quad (57)$$

for the general case (which will be needed in later dis-

cussion) in which the driving field energy is given by

$$E_D''' = E_D' e^{-at}. \quad (58)$$

The second term on the right side of Eq. (53), $\langle P_\gamma^2 \rangle$, is obtained easily from Eq. (39):

$$\langle P_\gamma^2 \rangle = (\omega^2 u^2 \gamma^2 / 4c^2) N [(e^{\alpha_0 t} - e^{\alpha_1 t}) / (\alpha_0 - \alpha_1)]^2. \quad (59)$$

For sufficiently large t this becomes

$$\langle P_\gamma^2 \rangle \approx (\omega^2 u^2 \gamma^2 / 4c^2) N (e^{2\alpha_0 t}) / (\alpha_0 - \alpha_1)^2. \quad (60)$$

The only term in Eq. (53) which has yet to be evaluated is $\langle P_F^2 \rangle$. From Eq. (40) we have

$$\langle P_F^2 \rangle = \int ds \int_0^\infty dt_1 \int_{-\infty}^{t_1} dt_2 \int ds' \int_0^\infty dt_1' \int_{-\infty}^{t_1'} dt_2' \times \mathcal{F}(t, s, t_1, t_2) \mathcal{F}(t, s', t_1', t_2') \frac{1}{2} \langle \{F(t_2), F(t_2')\} \rangle. \quad (61)$$

The symmetrized product in the integrand of Eq. (61) is obviously twice the symmetric part of the right side of Eq. (7). We have, therefore,

$$\frac{1}{2} \langle \{F(t_2), F(t_2')\} \rangle = \frac{\hbar \omega^3 \beta}{4\pi c^2} \delta(t_2 - t_2') f(T), \quad (62)$$

which we can also write as

$$\frac{1}{2} \langle \{F(t_2), F(t_2')\} \rangle = \frac{\hbar \omega^3 \beta}{8\pi^2 c^2} f(T) \int_{-\infty}^\infty d\nu e^{i\nu(t_2 - t_2')}. \quad (63)$$

Substituting in Eq. (61), we obtain

$$\langle P_F^2(t) \rangle = \frac{\hbar \omega^3 \beta}{8\pi^2 c^2} f(T) \int_{-\infty}^\infty d\nu Z(\nu) Z(-\nu), \quad (64)$$

where

$$Z(\nu) = \int ds \int_0^\infty dt_1 \int_{-\infty}^{t_1} dt_2 e^{i\nu t_2} \mathcal{F}(t, s, t_1, t_2). \quad (64a)$$

The evaluation of $Z(\nu)Z(-\nu)$, carried out in the Appendix, yields

$$\langle P_F^2(t) \rangle = \frac{\hbar \omega \beta}{4\pi^2 c^2} f(T) \int_{-\infty}^\infty dx \times \left| \frac{\alpha_0 \alpha_1}{\alpha_0 - \alpha_1} \frac{1}{2x + i\beta} \left(\frac{e^{\alpha_0 t}}{\alpha_0 + ix} - \frac{e^{\alpha_1 t}}{\alpha_1 + ix} \right) + \frac{x e^{-ixt}}{2(\alpha_0 + ix)(\alpha_1 + ix)} \right|^2. \quad (65)$$

Although the integration in Eq. (65) may be carried out explicitly, it is not worth the effort because our interest is mainly in those values of t for which the integrand can be considerably simplified. We note that for t sufficiently large, only the $e^{\alpha_0 t}$ term in the integrand contributes significantly to the integral. In that event

we have

$$\langle P_F^2(t) \rangle \approx \frac{\hbar\omega\beta}{16\pi^2c^2} f(T) \left(\frac{\alpha_0\alpha_1}{\alpha_0 - \alpha_1} \right)^2 \times e^{2\alpha_0 t} \int_{-\infty}^{\infty} \frac{dx}{(x^2 + \frac{1}{4}\beta^2)(x^2 + \alpha_0^2)}. \quad (66)$$

The integral in the above expression is $2\pi[\beta\alpha_0(\alpha_0 + \frac{1}{2}\beta)]^{-1}$. Making use of Eq. (26b), we have

$$\langle P_F^2(t) \rangle = \frac{\hbar\omega}{16\pi^2c^2} \frac{k^2}{(\alpha_0 - \alpha_1)^2} f(T) e^{2\alpha_0 t} \quad (67)$$

for sufficiently large t .

It will also be interesting to know the value of $\langle P_F^2(0) \rangle$. The expression within the absolute value signs in Eq. (65) becomes, for $t=0$,

$$\frac{x^2 + \frac{1}{2}i\beta x - \alpha_0\alpha_1}{2(\alpha_0 + ix)(\alpha_1 + ix)(x + \frac{1}{2}i\beta)} = \frac{-(\alpha_0 + ix)(\alpha_1 + ix)}{2(\alpha_0 + ix)(\alpha_1 + ix)(x + \frac{1}{2}i\beta)}, \quad (68)$$

so that

$$\langle P_F^2(0) \rangle = \frac{\hbar\omega\beta}{16\pi^2c^2} f(T) \int_{-\infty}^{\infty} \frac{dx}{(x^2 + \frac{1}{4}\beta^2)} = \frac{\hbar\omega}{8\pi^2c^2} f(T). \quad (69)$$

We have now all the terms in Eq. (53). We saw that $4\pi c^2 \langle P_D^2 \rangle_{av}$ is the energy of the amplified driving field, or the signal energy. The sum of the remaining two terms obviously represents spontaneous and thermally induced emission energy. It is incoherent (see I for a discussion of coherence)¹ and may be regarded as noise. We have, for $t \gg |\alpha_1|$,

$$4\pi c^2 \langle P_F^2 + P_F^2 \rangle \approx \frac{\hbar\omega k^2}{4(\alpha_0 - \alpha_1)^2 \langle I \rangle} [1 + \langle I \rangle f(T)] e^{2\alpha_0 t}, \quad (70)$$

where use has been made of Eq. (17c). This expression has the factor $[1 + \langle I \rangle f(T)]$, which is characteristic of spontaneous and thermally induced emission.³ In order to separate the two, we may set $T=0$ [yielding $f(T)=1$], thus obtaining an expression for spontaneous emission only. Subtracting the latter from the right side of Eq. (70) leaves thermally-induced emission only.

It is interesting to note the value which our formalism gives for the expectation value of the incoherent field at time $t=0$ (before the molecules begin to amplify). The same result is, of course, also obtained in the absence of molecules for arbitrary t . From Eqs. (59)

and (69) we have

$$\begin{aligned} 4\pi c^2 \langle P_F^2(0) + P_F^2(0) \rangle &= 4\pi c^2 \langle P_F^2(0) \rangle \\ &= \frac{1}{2} \hbar\omega f(T) \\ &= \frac{1}{2} \hbar\omega + \hbar\omega (e^{\hbar\omega/kT} - 1)^{-1}, \end{aligned}$$

which is just the sum of the zero-point energy and the thermal energy of the field, the latter being in accordance with Planck's radiation law.

Subsection 2

Let us consider spontaneous emission for the case $\langle I \rangle = 1$ (which means that the molecules are all in the upper state) in somewhat greater detail. It is given (for t sufficiently large) by

$$\langle H_f \rangle_{\text{spont}} \approx [\hbar\omega/2(\alpha_0 - \alpha_1)^2] k^2 e^{2\alpha_0 t}. \quad (71)$$

From Eq. (71) we can easily obtain the power radiated by the molecules spontaneously. Using the fact that the energy in the cavity comes mainly from the molecules, since the amplification is large, we have

$$-\frac{d}{dt} \sum_m \langle H_m \rangle = \beta \langle H_f \rangle + \frac{d}{dt} \langle H_f \rangle, \quad (72)$$

which states that the rate of increase in the field energy plus the power dissipated is equal to the power radiated by the molecules. This relationship may be applied separately to the induced- and spontaneous-emission fields (see II).¹ The spontaneous emission power radiated by the molecules is therefore given by

$$[-\alpha_1 k^2 / (\alpha_0 - \alpha_1)^2] e^{2\alpha_0 t}, \quad (73)$$

where use has been made of Eq. (26b). The total spontaneous-emission energy radiated by the molecules $E(t)$ is thus obtained, with some obvious approximations, as

$$E(t) = [\alpha_1^2 / (\alpha_0 - \alpha_1)^2] \hbar\omega e^{2\alpha_0 t}. \quad (74)$$

Equation (74) is an expression for the *minimum* energy which may be radiated by the molecules up to time t , namely, that which is radiated in the absence of a driving field. If we consider a situation where the signal energy is comparable to the spontaneous emission energy,⁸ so that the total energy radiated by the molecules is, e.g., $2E(t)$, then Eq. (74) gives us a method of determining the time during which the present analysis is valid. It was pointed out previously that this time must be less than that which produces a substantial change in the molecular properties. If the molecules are all initially in the upper state, for example, they will stop amplifying by the time they have lost half their energy, and some time before this the theory will have become invalid. If we require that

$$2E(t) < \epsilon N \hbar\omega, \quad (75)$$

⁸ This is often considered a limiting situation in amplifiers.

then ϵ might range, for instance, from $\frac{1}{10}$ to $\frac{1}{4}$ depending on whether we want a quantitative or merely qualitative description of the situation. Of course, if the expectation value of the initial energy of the molecules were only slightly above $\frac{1}{2}N\hbar\omega$, ϵ would have to be much smaller. Substituting from Eq. (74) into the inequality (75), and noting that the factor $\alpha_1^2/(\alpha_0 - \alpha_1)^2$ is of the order of unity (it varies from 1 in the limit of small k^2/β^2 to $\frac{1}{4}$ in the limit of large k^2/β^2), we have approximately

$$2\alpha_0 t < \ln(\epsilon N). \quad (76)$$

Thus, we obtain the upper limit on the time during which the theory is valid for the conditions considered.

Some of the expressions derived so far have assumed simpler form when we considered t "sufficiently large." It follows that this "sufficiently large" value⁷ must be much smaller than the upper limit indicated by the inequality (76); otherwise, the complete unsimplified expression must be used.

Subsection 3

A quantity which is always of interest is the ratio of induced- to spontaneous-emission energy in the cavity (which we will denote by R). We consider the situation for t sufficiently large, so that only the term containing the exponent with the positive exponential is significant. From Eqs. (54) and (71) we obtain, for the case of a constant driving field,

$$R' = E_D' 2\alpha_1^2 / \hbar\omega k^2. \quad (77)$$

We note first that this ratio is a constant, independent of the time, in contrast to the value obtained in II for a perturbation situation and exact molecular resonance.⁹ The factor $E_D'/\hbar\omega$ is obviously n , the number of photons in the driving field energy. We see (as has already been shown in II and III) that the ratio of induced to spontaneous emission is *not*, in general, equal to the number of photons in the driving-field energy. It is interesting to study the factor $2\alpha_1^2/k^2$ more closely. We have

$$2\alpha_1^2/k^2 = 1 + \frac{1}{4}\beta^2/k^2 + \frac{1}{4}\beta/k(\beta^2/k^2 + 8)^{1/2}. \quad (78)$$

It is seen that only in the limit of vanishing β/k or in the absence of cavity loss does the ratio of induced to spontaneous emission become equal to n . For the situation in which β/k is large compared to unity—which is the more usual case experimentally—the ratio of induced to spontaneous emission energy in the cavity is $\frac{1}{2}(\beta^2/k^2)n$ (for t sufficiently large). For the case in which the driving mechanism is shut off at $t=0$ and the driving field decays as $E_D'e^{-\beta t}$, we have from Eqs. (55) and (70)

$$R'' = E_D' 2\alpha_0^2 / \hbar\omega k^2 \quad (79)$$

⁹ Under perturbation conditions, which were discussed earlier, the spontaneous emission energy in the cavity approaches a steady-state value, while the induced emission energy does not. The latter is obtained immediately from the square of the expression in Eq. (45), and the former may be calculated from Eqs. (59) and (65).

for t sufficiently large,⁷ where E_D' is not the energy of the driving field but rather the energy which existed in the cavity when the driving mechanism was shut off. If the cavity loss is zero, $R'' = R' = n$, as would be expected. However, in the opposite limit, in which $k^2/\beta^2 \ll 1$, we have

$$R'' \approx E_D' 2k^2 / \hbar\omega\beta^2, \quad (80)$$

a quantity which is obviously much less than $E_D'/\hbar\omega$.

It is convenient at this point to refer to the work of Serber and Townes⁴ concerning fundamental limits of molecular amplification. Their argument rests on the supposition that the ratio of induced to spontaneous emission energy is n , which they derive without consideration of cavity losses. (If we let β approach zero, our results for induced and spontaneous emission become equivalent to theirs.) It can be seen, however, that this is not so in a realistic situation.

Thus far we have considered two types of driving field: one which is maintained constant by the driving mechanism, and one which decays freely with the driving mechanism shut off. A third type of driving field of interest is one for which the driving mechanism sends a constant amount of power into the cavity. In the absence of amplification the latter is equivalent to the constant driving field, but when the driving mechanism must interact with the amplified field, the two types of field are different.

The power transmitted to the field by the driving mechanism is given by

$$S = 4\pi c u_D \dot{D}(t) P(t). \quad (81)$$

We consider a driving dipole moment described by

$$D(t) = D_0 e^{-\frac{1}{2}at} \sin(\omega t + \theta), \quad (82)$$

where $a \ll \omega$. Then the driving field is

$$p_D(t) = [D_0 \omega u_D \cos(\omega t + \theta) / c(\beta - a)] (e^{-\frac{1}{2}at} - e^{-\frac{1}{2}\beta t}). \quad (83)$$

We assume that $a \ll \beta$, so that for $t \gg \beta$ we have

$$p_D(t) = p_{D0} \cos(\omega t + \theta) e^{-\frac{1}{2}at} = p_D'''(t), \quad (84)$$

namely, the driving field given in Eq. (35). $p_D(t)$ is therefore the field of Eq. (36) and the power delivered by the driving mechanism for t sufficiently large is obtained from expression (81) as

$$S = 2\pi c^2 p_{D0}^2 \beta [(-\alpha_0 \alpha_1) / (\alpha_0 - \alpha_1)] \times [e^{(\alpha_0 - \frac{1}{2}a)t} / (\alpha_0 + \frac{1}{2}a)]. \quad (85)$$

This will be a constant if

$$\frac{1}{2}a = \alpha_0. \quad (86)$$

We have thus the case of constant driving power by choosing a to satisfy Eq. (86). Since we have assumed that $a \ll \beta$, Eq. (86) implies that $\alpha_0 \ll \beta$, which in turn holds only if

$$k^2/\beta^2 \ll 1. \quad (87)$$

Equation (85) becomes

$$S = 2\pi c^2 p_{D0}^2 [(-\alpha_1)/2(\alpha_0 - \alpha_1)] \approx \pi c^2 p_{D0}^2 \beta. \quad (88)$$

The energy in the cavity is obtained from Eq. (57), for sufficiently large t , as

$$\begin{aligned} 4\pi c^2 \langle P_{D'''}^2 \rangle_{av} &= E_D' [\alpha_1/(\alpha_0 - \alpha_1)]^2 \frac{1}{4} e^{2\alpha_0 t} \\ &\approx \frac{1}{4} E_D' e^{2\alpha_0 t} \\ &\approx \frac{1}{2} (S/\beta) e^{2\alpha_0 t}. \end{aligned} \quad (89)$$

The ratio of induced to spontaneous emission energy is

$$R''' = \beta S / 4k^2 \hbar \omega. \quad (90)$$

At this point the following question arises: What power need be sent into the cavity to produce a signal equal to the spontaneous emission? Setting $R''' = 1$, we have

$$S = 4(k^2/\beta) \hbar \omega. \quad (91)$$

Equations (90) and (91) reveal an interesting fact. The amplification sensitivity (as measured either by S or by R''') increases as the cavity losses increase, although the amplification itself, of course, decreases. The obvious explanation for this fact is that an increase in cavity losses decreases the incoherent energy in the cavity more than it does the signal energy, as can be seen by comparing Eq. (70) with Eq. (89).

PART III

The consideration of a uniform molecular frequency spread is now in order. We are interested here in the same question as those considered in the case of exact molecular resonance. Since the ideas involved in the two analyses are similar, the treatment of the non-resonant case will be more concise than that of the resonant case.

A mathematically idealized situation is one in which the molecular density in frequency space, $\rho(\omega_m)$, is constant for all frequencies. It will be apparent from the calculations, however, that approximately the same results as those in the idealized situation are obtained for the case in which $\rho(\omega_m)$ is constant only in the neighborhood of the resonant frequency of the cavity ω , the neighborhood being somewhat larger than the inverse cavity relaxation time β (or cavity frequency width).

Our starting point is Eq. (14), where the summations of Eqs. (14a) and (14b) must now be carried out differently from the previous manner. Since the main contributions to the integrals in Eqs. (14a) and (14b) come from $\omega_m \sim \omega$ we can, firstly, drop terms in the integrand which oscillate with frequency $\omega_m + \omega$ when they are compared with those which oscillate with frequency $\omega_m - \omega$. We therefore have

$$p_\gamma = -\frac{\omega}{c} \sum_m u_m \begin{pmatrix} 0 & T_m^* \\ T_m & 0 \end{pmatrix}_m, \quad (92)$$

where

$$T_m = \frac{(e^{i\omega_m t} - e^{-\frac{1}{2}\beta t + i\omega t})}{[2(\omega - \omega_m) + i\beta]}. \quad (92a)$$

Also,

$$\begin{aligned} p &= -(8\pi/\hbar) \omega \tilde{\gamma}^2 \sum_m u_m^2 \langle I_m \rangle \int_0^t dt_2 \\ &\quad \times P(t_2) L(\omega_m - \omega, t - t_2), \end{aligned} \quad (93)$$

where

$$\begin{aligned} L(x, \tau) &\equiv -(x^2 + \frac{1}{4}\beta^2)^{-1} \\ &\quad \times [(\frac{1}{2}\beta \cos \omega \tau + x \sin \omega \tau)(\cos x \tau - e^{-\frac{1}{2}\beta \tau}) \\ &\quad + (x \cos \omega \tau - \frac{1}{2}\beta \sin \omega \tau) \sin x \tau]. \end{aligned} \quad (93a)$$

We carry out the summation over m in Eq. (93) by converting it into an integration over x . The first factor in Eq. (93a) shows that most of the contribution to the x integration comes from a neighborhood about $x=0$ which is of the order of β . We see, therefore, that this is the neighborhood in which $\rho(\omega_m)$ should be constant. We assume that $u_m^2 \langle I_m \rangle$ varies slowly in this neighborhood and replace it by its average $u^2 \langle I \rangle$. We then approximate the x integration by taking the limits to be $-\infty$ and $+\infty$. The odd terms in the integrand drop out and we have

$$p = b \int_0^t dt_2 P(t_2) e^{-\frac{1}{2}\beta(t-t_2)} \cos \omega(t-t_2), \quad (94)$$

where

$$b \equiv (4\pi^2/\hbar) \omega u^2 \tilde{\gamma}^2 \langle I \rangle \rho(\omega). \quad (94a)$$

Thus, there results another integral equation for P

$$P(t) = \psi(t) + \int_0^t dt_1 K(t-t_1) P(t_1) \quad (95)$$

of the same form as Eq. (17), but with the kernel now given by

$$K(t) = b e^{-\frac{1}{2}\beta t} \cos \omega t, \quad (95a)$$

and with

$$\psi(t) = p_F + p_D + p_\gamma, \quad (95b)$$

where p_F and p_D are still specified as previously [Eqs. (10a) and (10b)] but p_γ is now given by Eq. (92).

The integral equation (95) may be converted into a differential equation from which some results can be read off immediately. However, we will maintain a uniform procedure and obtain the solution directly from the integral equation by means of the Laplace transformation. This procedure involves no greater effort than that of obtaining all the necessary information from the differential equations and initial conditions. Since the method was explained in detail previously, only the results will be presented here. As in the case of the resonant molecules, the solution consists of three parts coming from the three terms in Eq. (95b)

$$P(t) = P_D + P_\gamma + P_F. \quad (96)$$

P_D is the induced field and depends on the type of driving field being considered. If we take as our driving field

$$p_D'''(t) = p_{D0} \cos(\omega t + \theta) e^{-\frac{1}{2}at} \equiv p_D'(t) e^{-\frac{1}{2}at}, \quad (97)$$

where

$$0 \leq a \ll \omega, \quad (97a)$$

then

$$P_D'''(t) = p_D'(t) \frac{be^{\frac{1}{2}(b-\beta)t} + (a-\beta)e^{-\frac{1}{2}at}}{(b-\beta+a)}. \quad (98)$$

From this general case of arbitrary a we obtain immediately two interesting special cases. Setting $a=0$, we have the case of a constant driving field $p_D'(t)$, resulting in

$$P_D'(t) = p_D'(t) \frac{be^{\frac{1}{2}(b-\beta)t} - \beta}{b-\beta}; \quad (99)$$

setting $a=\beta$, we have the case of a freely decaying driving field $p_D'(t)e^{-\frac{1}{2}\beta t}$ (the driving mechanism having been shut off at $t=0$), resulting in

$$P_D''(t) = p_D'(t) e^{\frac{1}{2}(b-\beta)t}. \quad (100)$$

The other two parts of the field are given by

$$P_\gamma = -\frac{\omega}{c} \tilde{\gamma} \sum_m u_m \begin{pmatrix} 0 & V_m^* \\ V_m & 0 \end{pmatrix}_m, \quad (101)$$

where

$$V_m = \frac{1}{2} e^{i\omega t} \frac{(e^{\frac{1}{2}(b-\beta)t} - e^{i(\omega_m - \omega)t})}{[(\omega_m - \omega) + \frac{1}{2}i(b-\beta)]}; \quad (101a)$$

and

$$P_F = \mathcal{L}^{-1}\{\bar{p}_F(s)/[1 - \bar{K}(s)]\}, \quad (102)$$

which reduces to Eq. (40) but with $\bar{K}(s)$ being the Laplace transform of the present kernel [Eq. (95a)]. The expectation value of P_F is zero for the same reasons as in the previous case, and the expectation value of P_γ —if there is no correlation between the phases of the molecules—is also zero. The expectation value of P_γ in the presence of molecular phase correlation may be derived easily but is of no present interest to us.

Equation (100) shows immediately that these molecules behave like a loss mechanism described by a loss constant $-b$ just as the true loss mechanism is described by the loss constant β . [This fact would have been evident immediately had we converted the integral Eq. (95) into a differential equation.] There is in this instance, however, the possibility of obtaining negative “loss” or gain, since b is positive when $\langle I \rangle$ is positive and $\langle I \rangle$ is positive when the molecules are mostly in the upper state. The fact that a nonresonant distribution of molecules acts like a linear loss mechanism is hardly surprising, since such a distribution was used as a specific model for a loss mechanism in II. At present, however, we are interested in the amplifying properties of the molecules and consider, therefore, only the case in which $\langle I \rangle$ and b are positive.

It is interesting to consider the situation of a constant driving field, described in Eq. (99), in some detail. For $b=0$ the field is, of course, equal to the driving field. For $0 < b < \beta$ the field approaches a steady-state value after a transient period which is of the order of $(b-\beta)^{-1}$. The steady-state value is larger than the driving field; the gain produced by the molecules reduces the loss, but not entirely. [For $b \ll \beta$ the situation is one to which perturbation theory may be applied, and it can be shown, with little difficulty, that the expression in Eq. (99) reduces to a special case of the perturbation theory expression in III.¹⁰] For $b=\beta$ the losses of the cavity are overcome completely, and an expansion of the exponential term shows that the field increases linearly with time, as though the cavity were lossless. Only for $b > \beta$ do we have an exponential amplification. This behavior is to be contrasted with that in the case of the resonant molecules, where there is eventual exponential amplification (subject to the restrictions inherent in our approximation method) no matter how small the gain.

The final subject under consideration is the energy. The signal energy is obtained immediately from the square of P (which should be averaged over a period and multiplied by $4\pi c^2$, as previously) P being given by Eqs. (98), (99), or (100), depending on the type of driving field we are considering. For a constant driving field and sufficiently large t [$t \gg (b-\beta)^{-1}$], the energy in the cavity is

$$E_D' [b/(b-\beta)]^2 e^{(b-\beta)t}, \quad (103)$$

where E_D' is the energy of the driving field, and in the case in which the driving mechanism is shut off at $t=0$, the energy in the cavity is

$$E_D' e^{(b-\beta)t}. \quad (104)$$

As previously, we are also interested in the situation in which the signal power sent into the cavity is constant. By the same arguments which were used in the corresponding case for the resonant molecules, we find that this situation results, again for sufficiently large t , if

$$a = b - \beta \quad (105)$$

and provided that

$$b - \beta \ll \beta. \quad (106)$$

The amplified signal energy in terms of the power S fed into the cavity is

$$(S/2\beta) [b/(b-\beta)] e^{(b-\beta)t}. \quad (107)$$

As far as the incoherent energy in the cavity is concerned, we have from Eq. (101), with a little calculation

$$\langle P_\gamma^2 \rangle = \frac{1}{2} \left(\frac{\omega u \tilde{\gamma}}{c} \right)^2 \rho(\omega) \frac{\pi}{b-\beta} [e^{(b-\beta)t} - 1], \quad (108)$$

¹⁰ The theory in III was developed for the distribution $\rho(\omega_m) = N\alpha\pi^{-\frac{1}{2}} \exp[-\alpha^2(\omega_m - \omega)^2]$. In order to convert the expressions in III into those for a uniform distribution ρ , one must substitute for N the quantity $\pi^{\frac{1}{2}}\rho/\alpha$ and then let α go to zero.

and from Eq. (102), by methods similar to those used previously in deriving Eq. (65),

$$\langle P_F^2 \rangle = \frac{\hbar\omega\beta}{16\pi^2c^2} f(T) \int_{-\infty}^{\infty} dx \times \frac{|(x + \frac{1}{2}i\beta)e^{ixt} + \frac{1}{2}ib e^{\frac{1}{2}(b-\beta)t}|^2}{(x^2 + \frac{1}{4}\beta^2)[x^2 + \frac{1}{4}(b-\beta)^2]}. \quad (109)$$

It is easy to verify that for $t=0$ or $b=0$, $\langle P_F^2 \rangle$ satisfies the same relationship [Eq. (69)] as that in the case of resonant molecules. For $t \gg (b-\beta)^{-1}$ we have for the incoherent (spontaneously emitted plus thermally induced) energy in the cavity,

$$4\pi c^2 \langle P_F^2 + P_T^2 \rangle = \frac{1}{2}\hbar\omega[b/(b-\beta)]\langle I \rangle^{-1}[1 + \langle I \rangle f(T)]e^{(b-\beta)t}. \quad (110)$$

If we set $T=0$ and $\langle I \rangle=1$, we have the simple expression for the spontaneous emission energy

$$\hbar\omega(b/b-\beta)e^{(b-\beta)t}. \quad (111)$$

We consider now the ratio R of signal energy to spontaneous-emission energy. For the case of constant driving field, Eqs. (103) and (111) yield

$$R' = E_D' b / \hbar\omega(b-\beta). \quad (112)$$

As shown previously in the case of the resonant molecules, this ratio equals the number of photons in the driving field *only* if the cavity is lossless. In the case of constant driving power, we have

$$R''' = \frac{1}{2}S/\beta\hbar\omega. \quad (113)$$

If we ask what the driving power must be in order that this ratio be unity, we obtain

$$S = 2\beta\hbar\omega.$$

Contrary to the situation in the case of the resonant molecules, we see that in this case the sensitivity decreases as the cavity losses increase, a result which causes no surprise.

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APPENDIX

We derive here Eq. (65) from Eq. (64). From Eqs. (40a) and (64a) we have

$$Z(\nu) = \frac{1}{\omega} \int_{\epsilon-i\infty}^{\epsilon+i\infty} ds \int_0^{\infty} dt_1 \int_{-\infty}^{t_1} dt_2 \times \frac{\exp[s(t-t_1) - \frac{1}{2}\beta(t_1-t_2)] \sin\omega(t_1-t_2)}{1-\bar{K}(s)}, \quad (A.1)$$

where the denominator of the integrand is given by Eq. (22). The t_2 and t_1 integrations may be carried out successively in a routine manner bearing in mind that the real part of s is positive. The result is

$$Z(\nu) = (1/\omega)B(\nu) \int ds \{[1-\bar{K}(s)](s-i\nu)\}^{-1}, \quad (A.2)$$

where

$$B(\nu) = [2(\nu-\omega)-i\beta]^{-1} - [2(\nu+\omega)-i\beta]^{-1}. \quad (A.3)$$

The integrand in Eq. (A.2) may be written as

$$\frac{(s^2+\omega^2)[(s+\frac{1}{2}\beta)^2+\omega^2]e^{st}}{[\varphi(s)(s-i\nu)]}, \quad (A.4)$$

where $\varphi(s)$ has the four roots given in Eqs. (26) and (26a). The integral in Eq. (A.2) is therefore given by

$$\sum_{j=1}^5 R_j(\nu), \quad (A.5)$$

where $R_j(\nu)$, $j=1, 2, 3, 4$, is the residue of (A.4) at the j th root of $\varphi(s)$, and R_5 is the residue at $s=i\nu$. These are given by

$$R_1(\nu) = \frac{Ae^{(i\omega+\alpha_0)t}}{i(\nu-\omega)-\alpha_0}, \quad (A.6)$$

$$R_2(\nu) = \frac{Ae^{(-i\omega+\alpha_0)t}}{i(\nu+\omega)-\alpha_0}, \quad (A.7)$$

$$R_3(\nu) = \frac{-Ae^{(i\omega+\alpha_1)t}}{i(\nu-\omega)-\alpha_1}, \quad (A.8)$$

$$R_4(\nu) = \frac{-Ae^{(-i\omega+\alpha_1)t}}{i(\nu+\omega)-\alpha_1}, \quad (A.9)$$

$$R_5(\nu) = \frac{(\omega^2-\nu^2)(\omega^2-\nu^2+i\nu\beta)e^{i\nu t}}{[i(\nu-\omega)-\alpha_0][i(\nu+\omega)-\alpha_0][i(\nu-\omega)-\alpha_1][i(\nu+\omega)-\alpha_1]}, \quad (A.10)$$

where

$$A \equiv \alpha_0 \alpha_1 / (\alpha_0 - \alpha_1).$$

One can see by inspection that

$$B(-\nu) = B^*(\nu), \quad \sum_j R_j(-\nu) = \sum_j R_j^*(\nu). \quad (\text{A.11})$$

We therefore have

$$\int_{-\infty}^{\infty} d\nu Z(\nu) Z(-\nu) = \int_{-\infty}^{\infty} d\nu |Z(\nu)|^2. \quad (\text{A.12})$$

It is easily seen that the main contribution to the integral (A.12) comes from the neighborhoods of $\nu = \pm\omega$, and the contributions from both neighborhoods are equal. Setting $x = \nu - \omega$ we have, in the neighborhood of $\nu = \omega$,

$$R_5 \approx \frac{x(2x - i\beta)e^{i(\omega+x)t}}{2(ix - \alpha_0)(ix - \alpha_1)}. \quad (\text{A.13})$$

Also, in the same neighborhood, R_2 and R_4 are negligible compared with R_1 and R_3 , and the second term in $B(\nu)$ is negligible compared to the first. We can thus write

$$Z(\nu)_{\nu \sim \omega} \approx \frac{e^{i\omega t}}{\omega} \left[\frac{A}{2x - i\beta} \left(\frac{e^{\alpha_0 t}}{ix - \alpha_0} - \frac{e^{\alpha_1 t}}{ix - \alpha_1} \right) - \frac{xe^{ixt}}{2(ix - \alpha_0)(ix - \alpha_1)} \right],$$

$$\equiv \tilde{Z}(x), \quad (\text{A.14})$$

and

$$\int_{-\infty}^{\infty} d\nu Z(\nu) Z(-\nu) = 2 \int_{-\infty}^{\infty} dx |\tilde{Z}(x)|^2. \quad (\text{A.15})$$

Substituting into Eq. (64), we obtain Eq. (65), with \tilde{Z} replaced by $-\tilde{Z}^*$.

Space-Charge Limited Current Relation in High-Pressure Gas Diodes

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A theory for space-charge conditions in high-pressure diodes has been developed which shows that the current obeys a $V^{1/2}$ power relation rather than the usual assumed V^2 relation. In addition, the theory predicts that the current in high-pressure diodes, at a constant anode voltage, varies as $p^{-1/2}$. Experimental data, taken in diodes filled with the inert gases, argon, neon, and helium, are presented to illustrate the validity of the theory.

THE Child-Langmuir¹ expression for space-charge conditions in a vacuum diode, showing that the current varies as the three-halves power of the voltage, has been verified by experiments over a wide range of conditions and been found to be true for any electrode that permits the escape of electrons or ions in a vacuum.

Although it is generally assumed that space-charge conditions apply to high-pressure gas diodes, there have been no widespread attempts to verify a relationship of this nature. The usual theoretical expression² which is considered applicable shows that the diode current follows a voltage-squared dependence given by

$$J = 9.95 \times 10^{-14} k V^2 / x^3 (\text{amp/cm}^2) \quad (1)$$

in the planar case and by

$$J = 0.56 \times 10^{-12} k V^2 / r^2 (\text{amp/cm}) \quad (2)$$

in cylindrical geometry. J is the current expressed in the units shown, k the mobility in $\text{cm}^2/\text{v sec}$, and x and r are anode distances in centimeters.

¹ I. Langmuir and K. T. Compton, *Revs. Modern Phys.* **3**, 191 (1931).

² J. D. Cobine, *Gaseous Conductors* (Dover Publications, Inc., New York, 1958), p. 129.

When space charge limited current measurements in high-pressure (1–300 mm) rare-gas diodes are made, they yield results which are at variance with the V^2 dependence of Eqs. (1) and (2). Figure 1 shows the

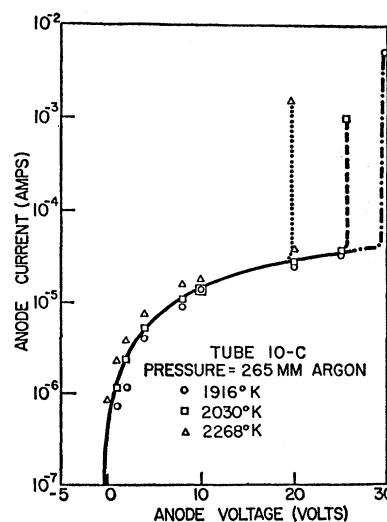


FIG. 1. Anode current-voltage characteristics of a cylindrical diode with a 0.009-in. diameter thoriated tungsten filament and a 1.0-cm diameter anode. The diode has an atmosphere of argon at 265 mm and the filament temperature is the other parameter.