

Induced and Spontaneous Emission in a Coherent Field. II

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The interaction between the electromagnetic field and a number of identical atomic systems, individually characterized by an electric dipole moment and two energy levels, is analyzed for the case where the atomic systems are inside a lossy cavity and exposed to a coherent driving field, resonance being assumed between atomic system, cavity, and driving field.

The problem of introducing loss into a quantum-mechanical formalism is treated first. Formal operator expressions are obtained for the field variables which include the absorption and the fluctuation (both thermal and quantum-mechanical) effects of the loss mechanism. Expectation values are then obtained for the field strength and the field energy which are valid for times short compared to the lifetime of the excited state. It is shown that the spontaneous-emission energy in the field increases initially as the square of the time and approaches a steady-state value after a transient period which is of the order of the cavity relaxation time. The induced emission contains two parts: incoherent emission

induced by the thermal field, and coherent emission induced by the driving field. The incoherent induced-emission energy has the same time dependence as the spontaneous-emission energy, and the ratio of the former to the latter is the number of photons in the thermal (inducing) field. The coherent induced-emission energy does not approach a steady-state value, but, after a transient period, increases linearly with the time. The ratio of the coherent induced-emission energy to the spontaneous-emission energy is equal, initially, to the number of photons in the driving-field energy, but becomes βt times as large after the transient period, where β is the reciprocal of the cavity relaxation time.

The expectation value of the rate of energy emission by the atomic systems is also obtained. It is shown that the ratio of the downward to the upward transition probabilities has the well-known value of $(n+1)/n$, where n is the field energy in units of the photon energy, *only* in the absence of a coherent field.

IN the first article of this series,¹ some of the problems peculiar to the interaction between a group of similar atomic systems and a coherent electromagnetic field were discussed. The atomic systems, referred to as molecules, were contained in a resonant cavity with perfectly conducting walls. The problems examined pertained mainly to the questions of coherence of the emission and the correlation of the molecular states. For these problems the idealization of a lossless cavity made possible a more illuminating discussion by eliminating complications which are not significant. There are other important problems, however, for which the cavity loss is of significance, such as the amplitudes of the induced- and spontaneous-emission fields in the cavity. Thus, for a constant-current radiating source inside a cavity, the field energy approaches a constant value if the cavity has some loss, while in the lossless case the energy increases indefinitely. For a realistic discussion, therefore, of the amount of spontaneous and induced emission and, in particular, of its time dependence (it is in this respect that interesting results are obtained), cavity loss must be considered. This is the purpose of the present article.

In the lossless case it was not necessary to include a driving mechanism to maintain a zero-order oscillating field in the cavity. Only an initial field was needed, since, because of the absence of cavity loss, this field continued to oscillate, in the lowest order, without change. (The interaction with the molecules is of higher order.) In the present case, however, an initial field will soon be damped out, so that a driving mechanism must be provided. We will, therefore, insert into the cavity, as our driving mechanism, a classical dipole of pre-

scribed dipole moment. The molecules, driving field, and cavity are all considered to be in resonance.

In Part I a method of introducing loss into a quantum-mechanical formalism is presented. Expectation values for the field strength and energy in the cavity in the presence of a driving field and a single molecule are derived and discussed in Part II. The power emitted by the molecule is considered in Part III, and expressions for the field due to a number of molecules are given in Part IV.

I

In order to explain as simply as possible the method of introducing loss into a quantum-mechanical formalism, we consider a cavity without molecules and without a driving mechanism; we have only the radiation field and the loss mechanism.

The effect of losses, or resistance, is the coupling of the electromagnetic field to the thermal bath, thereby converting electromagnetic energy into thermal energy. However, a coupling mechanism works in either direction. It will, therefore, transmit thermal (and also, as will be seen, quantum-mechanical) fluctuations to the electromagnetic field.

In order to have a loss mechanism which behaves like a resistance, we impose the following two requirements: (1) The loss mechanism must be nonresonant. (2) The state of the loss mechanism is determined mainly by the temperature of the thermal bath to which it is coupled, and only slightly by the field in the cavity. The first requirement may be met by considering the losses to be due to a large number of electric dipoles, the resonant frequencies of which are continuously distributed. The second requirement will be considered later.

¹I. R. Senitzky, Phys. Rev. **111**, 3 (1958), hereafter referred to as I.

We consider, therefore, a cavity with walls which are still perfectly reflecting, but include inside the cavity a large number of dipoles, each one labeled with index "j" and having (angular) frequency ω_j . For simplicity we take each dipole to be of the type studied in I, that is, having only two energy states, their difference being $\hbar\omega_j$, and having no permanent dipole moment; but now there are dipoles within the cavity for all (positive) values of ω_j . The number of dipoles per unit frequency range (dipole density) is $\rho(\omega_j)$, which is a slowly varying function of ω_j . The notation used² will be essentially the same as that in I, except that the dipole moment operator of the dipoles constituting the loss mechanism will be designated by Γ , the symbol γ , used in I, being reserved for the molecules to be introduced into the cavity later.

The Hamiltonian for the system of cavity field and dipoles is

$$H = H_{\text{field}} + \sum_j H_j + 4\pi c P \sum_j \Gamma_j \cdot \mathbf{u}(\mathbf{r}_j), \quad (1)$$

where H_j is the Hamiltonian of the j th dipole containing its internal variables, \mathbf{r}_j is its position in the cavity considered to be fixed, and

$$H_{\text{field}} = 2\pi c^2 P^2 + (\omega^2/8\pi c^2) Q^2. \quad (2)$$

We have not indicated explicitly any coupling of the dipoles to the thermal bath, and this requires some explanation. If the number of dipoles were small, then, in order for the loss mechanism to absorb a given power from the field and still satisfy the second requirement (i.e., that its state be determined mainly by the temperature of the thermal bath), the dipoles would have to be coupled tightly to the thermal bath so that they would transmit the energy to the bath almost as soon as they received it from the field. If, however, we take the number of dipoles to be very large, then the coupling to the thermal bath may be taken to be weak; and if the period of time under consideration is not excessively large, this coupling may be eliminated entirely. In the latter situation we merely require that the *initial* state of the mechanism be determined by a temperature; the very large heat capacity of the total number of dipoles makes it possible to satisfy the second requirement. Our treatment will refer to this idealized situation, in which the effect of the coupling to the thermal bath on the motion of the dipoles may be neglected.

For simplicity we consider $\mathbf{u}(\mathbf{r}_j)$ to have the same value \mathbf{U} for all j , and designate by Γ_j the component of Γ_j along \mathbf{U} . The equation of motion for Γ_j is

$$i\hbar d\Gamma_j/dt = [\Gamma_j(t), H_j(t)], \quad (3)$$

which, together with

$$H_j = H_j(0) + \frac{4\pi c}{i\hbar} U \int_0^t dt_1 [H_j(t_1), \Gamma_j(t_1)] P(t_1), \quad (4)$$

² The field is described by $\mathbf{E} = -4\pi c \mathbf{P}$, $\mathbf{H} = \nabla \times \mathbf{A}$, $\mathbf{A} = Q(t) \mathbf{u}(\mathbf{r})$, $\mathbf{P} = P(t) \mathbf{u}(\mathbf{r})$, where $[Q(t), P(t)] = i\hbar$, and $\mathbf{u}(\mathbf{r})$ is a normalized function describing the spatial dependence of the field for the mode under consideration, the normalization being $\int_V u^2(\mathbf{r}) d^3\mathbf{r} = 1$, where V is the volume of the cavity.

gives

$$\frac{d\Gamma_j(t)}{dt} = \frac{1}{i\hbar} [\Gamma_j(t), H_j(0)] + \frac{4\pi c}{\hbar^2} \int_0^t dt_1 [\Gamma_j(t), [\Gamma_j(t_1), H_j(t_1)] P(t_1)]. \quad (5)$$

As stated in our second requirement concerning the loss mechanism, the effect of the field on each dipole is small. We may, therefore, approximate Γ_j and H_j under the integral sign in Eq. (5) by $\Gamma_j^{(0)}$ and $H_j^{(0)}$, which are the corresponding operators for the dipole uncoupled from—or unperturbed by—the field.³ We approximate further by neglecting $[P(t_1), \Gamma_j^{(0)}(t)]$ compared to $\Gamma_j^{(0)}(t) P(t_1)$ under the integral sign.⁴ The first term on the right side of Eq. (5) becomes $d\Gamma_j^{(0)}(t)/dt$ [see discussion following Eq. (I, 12)]. We thus obtain

$$\Gamma_j(t) = \Gamma_j^{(0)}(t) + \frac{4\pi c}{\hbar^2} U \times \int_0^t dt_1 \int_0^{t_1} dt_2 [\Gamma_j^{(0)}(t_1), [\Gamma_j^{(0)}(t_2), H_j^{(0)}]] P(t_2). \quad (6)$$

Our treatment of the dipole resembles perturbation theory. Thus, we obtain Eq. (6) from Eq. (5) by considering only the lowest-order interaction in the expression for the dipole moment. It should be borne in mind, however, that our treatment of the field must not be a perturbation treatment, since we are dealing with a large number of dipoles, and their effect on the field may be great. In fact, as will be seen, the effect on the field is described by the product of dipole density and dipole moment, and this product need not be small even though the dipole moment itself is small.

It is interesting to note that the substitution of the unperturbed dipole operators under the integral sign in Eq. (5) has the effect of making the dissipative system linear. The perturbed operators Γ_j and H_j contain, in general, a dependence on P , so that the dipole current is a nonlinear function of the field. However, $\Gamma_j^{(0)}$ and $H_j^{(0)}$ are independent of P , and their substitution results in a linear dependence of the current on the field.

Denoting the off-diagonal matrix elements of $\Gamma_j^{(0)}$ by $\bar{\Gamma}_j$,³ and making use of the relationship [see equation

$$H_j^{(0)} = \begin{pmatrix} E_1(j) & 0 \\ 0 & E_2(j) \end{pmatrix}, \quad \Gamma_j^{(0)}(t) = \bar{\Gamma} \begin{pmatrix} 0 & e^{-i\omega_j t} \\ e^{i\omega_j t} & 0 \end{pmatrix},$$

where $E_2(j) - E_1(j) = \hbar\omega_j$. Also, see discussion following Eq. (I, 12).

³ Explicitly, in the energy representation of the free dipoles, we have (in accordance with I)

⁴ Since $\Gamma^{(0)}(t)$ is uncoupled from the field, one might at first glance expect that it commutes within the field. The latter, however, is not uncoupled from the dipoles, and its description contains $\Gamma^{(0)}(t')$, $t' \neq t$. We can obtain a rough estimate of the neglected commutator by using Eq. (21) to evaluate it. It can be shown that the approximation is justified for our purposes.

preceding Eq. (I, 13)

$$[\Gamma_j^{(0)}(t), [\Gamma_j^{(0)}(t_1), H_j^{(0)}]] = 2\hbar\omega_j \bar{\Gamma}^2 I_j \cos\omega_j(t-t_1), \quad (7)$$

where

$$I_j \equiv \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}$$

for the j th dipole and unit matrix for the other dipoles, we obtain

$$\Gamma_j(t) = \Gamma_j^{(0)}(t) + \frac{8\pi c}{\hbar} U \bar{\Gamma}^2 I_j \int_0^t dt_1 P(t_1) \sin\omega_j(t-t_1). \quad (8)$$

This is an operator equation, $\Gamma_j^{(0)}$ and I_j being operators with respect to the dipole coordinates, and P being an operator with respect to the field coordinates. For convenience in later calculation, we make an approximation at this point, replacing I_j by its expectation value (times the unit operator), $\langle I_j \rangle$. Since our end results will be expectation values anyway, this approximation only eliminates, in effect, higher order quantum-mechanical interaction terms than the second (which are related to correlation effects between dipole and field fluctuations), and is consistent with the approximation previously made with respect to the dipole moment.

Utilizing the expressions which describe the field in terms of its initial value and the dipole moments, namely [see Eq. (I, 5)],

$$Q(t) = Q^{(0)}(t) + 4\pi c \sum_j \int_0^t dt_1 \Gamma_j(t_1) U \cos\omega(t-t_1), \quad (9)$$

$$P(t) = P^{(0)}(t) - \frac{\omega}{c} \sum_j \int_0^t dt_1 \Gamma_j(t_1) U \sin\omega(t-t_1),$$

where

$$\begin{aligned} P^{(0)}(t) &= P(0) \cos\omega t - (\omega/4\pi c^2) Q(0) \sin\omega t, \\ Q^{(0)}(t) &= Q(0) \cos\omega t + (4\pi c^2/\omega) P(0) \sin\omega t, \end{aligned} \quad (10)$$

we obtain

$$\begin{aligned} P(t) &= P^{(0)}(t) - \frac{\omega}{c} U \sum_j \int_0^t dt_1 \Gamma_j^{(0)}(t_1) \sin\omega(t-t_1) \\ &\quad - \frac{8\pi\omega}{\hbar} U^2 \bar{\Gamma}^2 \sum_j \langle I_j \rangle \int_0^t dt_1 \int_0^{t_1} dt_2 P(t_2) \\ &\quad \times \sin\omega_j(t_1-t_2) \sin\omega(t-t_1), \end{aligned} \quad (11)$$

and

$$\begin{aligned} Q(t) &= Q^{(0)}(t) + 4\pi c \sum_j \int_0^t dt_1 \Gamma_j^{(0)}(t_1) \cos\omega(t-t_1) \\ &\quad + \frac{32\pi^2 c^2}{\hbar} U^2 \bar{\Gamma}^2 \sum_j \langle I_j \rangle \int_0^t dt_1 \int_0^{t_1} dt_2 P(t_2) \\ &\quad \times \sin\omega_j(t_1-t_2) \cos\omega(t-t_1). \end{aligned} \quad (12)$$

We carry out the t_1 integration in the last term of Eq. (11). Noting that the significant contribution to the integral (for $t \gg \omega^{-1}$) will arise from the $\omega_j \sim \omega$ terms, we ignore the $(\omega_j + \omega)^{-1}$ terms compared to the $(\omega_j - \omega)^{-1}$ terms. The result for the last term in Eq. (11) is

$$\begin{aligned} &\frac{8\pi\omega}{\hbar} U^2 \bar{\Gamma}^2 \sum_j \langle I_j \rangle \frac{1}{\omega_j - \omega} \int_0^t dt_1 P(t_1) \\ &\quad \times \sin \frac{1}{2}(\omega_j - \omega)(t-t_1) \cos \frac{1}{2}(\omega_j + \omega)(t-t_1). \end{aligned} \quad (13)$$

We can now carry out the summation by converting it into an integration, making the usual approximations which go with the fact that the main contributions to (13) come from the terms for which $\omega_j \sim \omega$ (again for $t \gg \omega^{-1}$). We obtain, thus,

$$\begin{aligned} P(t) &= P^{(0)}(t) - \frac{\omega}{c} U \sum_j \int_0^t dt_1 \Gamma_j^{(0)}(t_1) \sin\omega(t-t_1) \\ &\quad - \beta \int_0^t dt_1 P(t_1) \cos\omega(t-t_1), \end{aligned} \quad (14)$$

where

$$\beta \equiv - (4\pi^2\omega/\hbar) U^2 \bar{\Gamma}^2 \langle I_j \rangle_{\omega} \rho(\omega), \quad (15)$$

$\langle I_j \rangle_{\omega}$ being the average expectation value⁵ of I_j for those dipoles the frequency of which is in the neighborhood of ω . Since $P^{(0)}(t)$ and $\Gamma^{(0)}(t)$ are known operators, Eq. (14) is an integral equation for $P(t)$. We can cast it into more familiar form by transforming it into a differential equation. Remembering that

$$\dot{P}^{(0)} + \omega^2 P^{(0)} = 0, \quad (16)$$

we have

$$\dot{P} + \beta \dot{P} + \omega^2 P = - (\omega^2/c) U \sum_j \Gamma_j^{(0)}(t). \quad (17)$$

It is worthwhile noting certain interesting features of Eq. (17). In the absence of a loss mechanism, the field satisfies Eq. (16). If we were to include cavity losses phenomenologically, we would most likely only add the $\beta \dot{P}$ term to the left side of Eq. (16), leaving the right side zero. Then we would have an operator equation in quantum mechanics identical to the dynamical variable equation in classical mechanics—a common situation. However, the term on the right side of Eq. (17) is very important, even if one ignores thermal fluctuations, since, as will be seen in the subsequent analysis, it accounts for quantum fluctuations which are formally responsible for spontaneous emission.

The initial conditions contained in Eq. (14) are expressed in terms of $P(0)$ and $Q(0)$. In connection with Eq. (17), we need initial conditions in terms of $P(0)$ and $\dot{P}(0)$, the latter, as obtained from Eq. (14), being

$$\dot{P}(0) = - (\omega^2/4\pi c^2) Q(0) - \beta P(0). \quad (18)$$

We can now write down the solution of Eq. (17). The

⁵ It is assumed that $\langle I_j \rangle$ is a slowly varying function of ω_j .

solution of the homogeneous equation satisfying the initial conditions is

$$e^{-\frac{1}{2}\beta t} \left\{ P(0) \cos \omega' t - \frac{1}{\omega'} \left[\frac{\omega^2}{4\pi c^2} Q(0) + \frac{1}{2} \beta P(0) \right] \sin \omega' t \right\}, \quad (19)$$

and a particular solution which is zero initially is

$$-\frac{\omega^2}{\omega' c} U \sum_j \int_0^t dt_1 \Gamma_j^{(0)}(t_2) e^{-\frac{1}{2}\beta(t-t_1)} \sin \omega'(t-t_1), \quad (20)$$

where

$$\omega' \equiv \omega [1 - (\beta/2\omega)^2]^{\frac{1}{2}}.$$

We assume that $\beta/\omega \ll 1$. We can, therefore, write, as the approximate expression for the complete solution,

$$P(t) = e^{-\frac{1}{2}\beta t} P^{(0)}(t) - \frac{\omega}{c} U \sum_j \int_0^t dt_1 \Gamma_j^{(0)}(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin \omega(t-t_1). \quad (21)$$

In an entirely analogous manner, we obtain

$$Q(t) = e^{-\frac{1}{2}\beta t} Q^{(0)}(t) + 4\pi c U \sum_j \int_0^t dt_1 \Gamma_j^{(0)}(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \cos \omega(t-t_1). \quad (22)$$

Equations (21) and (22) are the explicit expressions for the operators P and Q in terms of the initial operators $P(0)$, $Q(0)$, and $\Gamma_j(0)$.

It is easy to justify these expressions intuitively. The resistance in the cavity acts as an absorber and an emitter. The absorptive properties account for the factors containing β , and the emission accounts for the second term in both expressions.

To illustrate the significance of these expressions, we solve two simple problems, the solutions of which are well known.⁶ The insight gained thereby will be helpful in the subsequent analysis of more complicated situations. We will obtain the expectation values for the field strength and for the energy.

In order to obtain expectation values, we must describe the initial state of our system. We take the field to be oscillating, initially, with amplitude $E_0 \mathbf{u}(\mathbf{r})$, in the manner described in I. The initial state of the j th dipole is described by $a_1(j) \varphi_1(j) + a_2(j) \varphi_2(j)$, where $\varphi_1(j)$ and $\varphi_2(j)$ are the lower and upper energy states, respectively, and

$$|a_1|^2 + |a_2|^2 = 1. \quad (23)$$

We consider the dipoles to be, initially, in equilibrium with a thermal bath at temperature T . In accordance with the principles of statistical mechanics, the phases of $a_1(j)$ and $a_2(j)$ are random, and we may set

$$|a_2(j)|^2 / |a_1(j)|^2 = \exp(-\hbar \omega_j / kT). \quad (24)$$

⁶ See H. B. Callen and T. A. Welton, Phys. Rev. **83**, 34 (1951), whose methods are applied by J. Weber, Phys. Rev. **90**, 977 (1953).

Since

$$\langle \Gamma_j^{(0)} \rangle = 2 |a_1(j) a_2(j)| \bar{\Gamma} \cos(\omega_j t + \theta_j),$$

where θ_j is the phase difference between $a_2(j)$ and $a_1(j)$ [see Eq. (I, 10)], and since θ_j is a random variable,

$$\sum_j \langle \Gamma_j^{(0)}(t) \rangle = 0, \quad (25)$$

so that

$$\langle \mathbf{E}(\mathbf{r}) \rangle = -4\pi c \mathbf{u}(\mathbf{r}) \langle P(t) \rangle = E_0 \mathbf{u}(\mathbf{r}) e^{-\frac{1}{2}\beta t} \sin \omega t. \quad (26)$$

The amplitude of the oscillations decays exponentially, and we have the same expression for the expectation value of the field as that which would be obtained for a classical field if we set

$$\beta = \omega / "Q", \quad (27)$$

where "Q" is the quality factor of the cavity.⁷

To obtain the expectation value of the energy of the field, we evaluate

$$\langle H_{\text{field}} \rangle = 2\pi c^2 \langle P^2 \rangle + (\omega^2 / 8\pi c^2) \langle Q^2 \rangle. \quad (28)$$

Now, the expectation value of all cross terms in P^2 and Q^2 [see Eqs. (21) and (22)] vanishes, since

$$\sum_j \langle P^{(0)} \Gamma_j \rangle = \langle P^{(0)} \rangle \sum_j \langle \Gamma_j^{(0)} \rangle = 0, \quad (29)$$

and

$$\sum_{j \neq k} \langle \Gamma_j^{(0)} \Gamma_k^{(0)} \rangle = \sum_j \langle \Gamma_j^{(0)} \rangle \sum_k \langle \Gamma_k^{(0)} \rangle = 0.$$

The value of

$$2\pi c^2 \langle P^{(0)2} \rangle + (\omega^2 / 8\pi c^2) \langle Q^{(0)2} \rangle$$

is the expectation value of the initial energy of the cavity field and, according to Eq. (I, 18), is given by $E_0^2 / 8\pi + \frac{1}{2} \hbar \omega$. We are thus left with only the evaluation of

$$\frac{\omega^2}{c^2} U^2 \sum_j \int_0^t dt_1 \int_0^t dt_2 \langle \Gamma_j^{(0)}(t_2) \Gamma_j^{(0)}(t_1) \rangle e^{-\frac{1}{2}\beta(2t-t_1-t_2)} \times \sin \omega(t-t_1) \sin \omega(t-t_2), \quad (30)$$

for P^2 and the corresponding term for Q^2 . Noting from the equation following Eq. (I, 17) that

$$\langle \Gamma_j^{(0)}(t_2) \Gamma_j^{(0)}(t_1) \rangle = \bar{\Gamma}^2 (|a_1|^2 e^{i\omega_j(t_1-t_2)} + |a_2|^2 e^{-i\omega_j(t_1-t_2)}), \quad (31)$$

we obtain for expression (30)

$$\frac{\omega^2}{c^2} U^2 \bar{\Gamma}^2 \int_0^t dt_1 \int_0^t dt_2 e^{-\frac{1}{2}\beta(2t-t_1-t_2)} \times \sin \omega(t-t_1) \sin \omega(t-t_2) \sum_j e^{i\omega_j(t_1-t_2)}, \quad (32)$$

which is shown in Appendix A to be equal to

$$\frac{\omega^2}{c^2} U^2 \bar{\Gamma}^2 \rho(\omega) \int_0^t dt_1 e^{-\beta(t-t_1)} \sin^2 \omega(t-t_1). \quad (33)$$

⁷ In order to distinguish the quality factor from the coordinate of the cavity field, we use quotation marks about the former.

The corresponding term for Q^2 is

$$16\pi^3 c^2 U^2 \bar{\Gamma}^2 \rho(\omega) \int_0^t dt_1 e^{-\beta(t-t_1)} \cos^2 \omega(t-t_1). \quad (34)$$

Multiplying (33) by $2\pi c^2$, (34) by $\omega^2/8\pi c^2$, and adding the results, we obtain, as the part of the energy due to the dipoles,

$$\frac{1}{2} \hbar \omega (1 - e^{-\beta t}) (-\langle I_j \rangle_\omega)^{-1}, \quad (35)$$

where we have made use of Eq. (15). Since

$$\langle I_j \rangle = |a_2(j)|^2 - |a_1(j)|^2,$$

we obtain from Eqs. (23) and (24)

$$(-\langle I_j \rangle_\omega)^{-1} = 1 + 2[\exp(\hbar\omega/kT) - 1]^{-1}. \quad (36)$$

We have, therefore, as the part of the field energy which comes from the initial field,

$$(E_0^2/8\pi + \frac{1}{2} \hbar \omega) e^{-\beta t}, \quad (37)$$

and, as the part of the field energy which comes from the dipoles,

$$\frac{1}{2} \hbar \omega (1 - e^{-\beta t}) + \hbar \omega (e^{\hbar\omega/kT} - 1)^{-1} (1 - e^{-\beta t}). \quad (38)$$

We see that the initial classical energy, $E_0^2/8\pi$, decays as $e^{-\beta t}$; that is, at the classical rate. It is interesting to note that the initial zero-point energy in (37) also decays at the same rate, but that the zero-point energy due to the dipoles, in (38), increases at a compensating rate to keep the total constant, so that at absolute zero

$$\langle H_{\text{field}} \rangle_{T=0} = (E_0^2/8\pi) e^{-\beta t} + \frac{1}{2} \hbar \omega. \quad (39)$$

For $\beta t \gg 1$ the zero-point energy is due entirely to the dipoles. This illustrates, in part, the significance of the right member of Eq. (17), or the second terms on the right sides of Eqs. (21) and (22). These terms also account for the second term in (38), which for $T > 0$ gives the thermal field energy. Since it was assumed that at $t=0$ there is no thermal field energy in the cavity, this energy builds up from zero to a steady-state value

$$\hbar \omega (e^{\hbar\omega/kT} - 1)^{-1}, \quad (40)$$

which is in agreement with Planck's radiation law.

Perhaps the most fundamental justification for the second term on the right sides of Eqs. (21) and (22), from a quantum-mechanical viewpoint, is the commutation relationship between Q and P . The contribution of the first term towards $[Q, P]$ is $i\hbar e^{-\beta t}$, which, standing alone, would obviously result in a violation of the uncertainty principle. It is shown in Appendix B that the contribution of the second term toward the commutator is $i\hbar(1 - e^{-\beta t})$, which is just what is needed to produce the correct commutation relationship, $[Q, P] = i\hbar$.

II

We come now to the central problem, namely, the consideration of the behavior of molecules in a lossy

cavity in the presence of a driving field. For simplicity we consider, at first, the presence of only a single molecule inside the cavity. This molecule is the same as that considered in I. Its internal energy is described by a Hamiltonian H_m , which, for the free molecule (uncoupled from the cavity field), has the eigenvalues E_1 and E_2 , with $E_2 - E_1 = \hbar\omega$, and the eigenstates φ_1 and φ_2 . The position of the molecule in the cavity is \mathbf{r}_m , and its dipole moment is γ_m . The component of dipole moment along $\mathbf{u}(\mathbf{r}_m)$ is designated by γ_m . For the free molecule, $\gamma_m(0)$ has the off-diagonal matrix elements $\tilde{\gamma}$ and the diagonal matrix elements zero. The initial state of the molecules is specified by $a_1\varphi_1 + a_2\varphi_2$, where $|a_1|^2 + |a_2|^2 = 1$. The phase difference between a_2 and a_1 will be designated by θ_m .

For the driving mechanism we consider a classical dipole of prescribed (oscillating) electric dipole moment

$$D(t) \equiv D_0 \sin(\omega t + \theta)$$

situated at \mathbf{r}_D . Instead of Eq. (1), we now have for the Hamiltonian

$$H = H_{\text{field}} + \sum_j H_j + H_m + 4\pi c P (\sum_j \Gamma_j U + \gamma_m u + D u_D), \quad (41)$$

where u_D is the component of $\mathbf{u}(\mathbf{r}_D)$ along the driving dipole. In accordance with Eq. (I, 5) we must add to the right side of Eq. (11) the term

$$-\frac{\omega}{c} \int_0^t dt_1 [\gamma_m(t_1) u + D(t_1) u_D] \sin \omega(t - t_1).$$

We notice that this has the same form as the $\Gamma_j^{(0)}$ terms in Eq. (11), and may be included in the subsequent analysis leading to Eq. (21) merely by adding $u\gamma_m(t_1) + u_D D(t_1)$ to $\sum_j U\Gamma_j^{(0)}$, so that instead of Eq. (21) we now have

$$P(t) = e^{-\frac{1}{2}\beta t} P^{(0)}(t) - \frac{\omega}{c} \int_0^t dt_1 e^{-\frac{1}{2}\beta(t-t_1)} [\sum_j U\Gamma_j^{(0)}(t_1) + u\gamma_m(t_1) + u_D D(t_1)] \sin \omega(t - t_1). \quad (42)$$

In a similar manner, Eq. (22) is replaced by

$$Q(t) = e^{-\frac{1}{2}\beta t} Q^{(0)}(t) + 4\pi c \int_0^t dt_1 e^{-\frac{1}{2}\beta(t-t_1)} [\sum_j U\Gamma_j^{(0)}(t_1) + u\gamma_m(t_1) + u_D D(t_1)] \cos \omega(t - t_1). \quad (43)$$

Equations (42) and (43) are no longer explicit expressions for the operators P and Q in the manner of Eqs. (21) and (22), since γ_m is now one of the three dynamical variables, P , Q , and γ_m , which are to be obtained from the equations of motion. Equations (42) and (43) are two of these equations, and the third is the same as Eq. (I, 9):

$$i\hbar \dot{\gamma}_m(t) = [\gamma_m(t), H_m(t)]. \quad (44)$$

We proceed now to solve these equations by means of perturbation theory along the same lines as those in I.

The new feature is the inclusion of losses and a classical driving field. It should be pointed out that only the interaction between molecule and field is treated by perturbation theory. The other interactions are treated more precisely.

There is good reason for the use of perturbation theory (besides the fact that it offers a method of solving the problem) in the analysis of the interaction between molecule and field. The processes of spontaneous emission, induced emission, and absorption have meaning only within a perturbation-theory—or small-signal—framework. Outside this framework these processes become intertwined and inseparable, and a discussion of them individually, which is very helpful in an understanding of the interaction, is impossible. The limitations of perturbation theory, of course, are well known. In our case the theory is valid only for times short compared to the lifetimes of the initial states; that is, for times short compared to the time required for the expectation value of the molecular energy to change substantially. This time depends, of course, on the strength of the cavity field. For fields which are not too strong, it will be much longer than the cavity relaxation time, β^{-1} . In the following discussion, the time is to be understood as properly restricted in accordance with the above considerations.

We now expand P , Q , and γ_m in powers of the coupling constant between molecule and field (considered to be included in $\tilde{\gamma}$) in the following manner:

$$P = P^{[0]} + P^{[1]} + P^{[2]} + \dots \quad (45)$$

It is important to distinguish between $P^{[0]}$ and $P^{(0)}$ (and likewise for the higher orders). $P^{(0)}$ describes the cavity field uncoupled to the loss mechanism, the driving mechanism, and the molecule; while $P^{[0]}$ describes the cavity field coupled to the loss mechanism and the driving mechanism (to all orders) but uncoupled to the molecules.

In setting up the equation of motion, we have considered the initial time to be $t=0$, and the initial conditions refer to this time. For our present purposes, it is more convenient to take the initial time to be $-\infty$. This has the effect of eliminating the $P^{(0)}$ and $Q^{(0)}$ terms in Eqs. (42) and (43), and changing the lower limit of the integrations to $-\infty$. Physically, these modifications mean that the initial oscillating field and the transient part of the driving field have been damped out before the time under consideration. Now, we want the molecule to enter the cavity at $t=0$. Mathematically, this is accomplished if we consider the position of the molecule, \mathbf{r}_m , to be a function of the time, and

$$\begin{aligned} \mathbf{u}[\mathbf{r}_m(t)] &= 0, & t < 0; \\ \mathbf{u}[\mathbf{r}_m(t)] &= \mathbf{u}, & t > 0. \end{aligned} \quad (46)$$

We have then

$$P(t) = P_D + P_\Gamma + P_\gamma, \quad (47)$$

$$Q(t) = Q_D + Q_\Gamma + Q_\gamma, \quad (48)$$

where

$$P_D = \frac{\omega u_D}{c\beta} D_0 \cos(\omega t + \theta) \equiv P_{D0} \cos(\omega t + \theta),$$

$$Q_D = \frac{4\pi c u_D}{\beta} D_0 \sin(\omega t + \theta) \equiv Q_{D0} \sin(\omega t + \theta),$$

$$P_\Gamma = -\frac{\omega}{c} U \sum_j \int_{-\infty}^t dt_1 \Gamma_j^{(0)}(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1),$$

$$Q_\Gamma = 4\pi c U \sum_j \int_{-\infty}^t dt_1 \Gamma_j^{(0)}(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \cos\omega(t-t_1),$$

$$P_\gamma = -\frac{\omega}{c} u \int_0^t dt_1 \gamma_m(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1),$$

$$Q_\gamma = 4\pi c u \int_0^t dt_1 \gamma_m(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \cos\omega(t-t_1).$$

The zeroth order expressions for $P(t)$ and $Q(t)$ may be written down immediately. We have

$$\begin{aligned} P^{[0]}(t) &= P_D(t) + P_\Gamma(t), \\ Q^{[0]}(t) &= Q_D(t) + Q_\Gamma(t). \end{aligned} \quad (49)$$

It is obvious from Eq. (44) that $\gamma_m^{[0]} = \gamma_m^{(0)}$. We can, therefore, write

$$P^{[1]} = -\frac{\omega}{c} u \int_0^t dt_1 \gamma_m^{(0)}(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1), \quad (50)$$

$$Q^{[1]} = 4\pi c u \int_0^t dt_1 \gamma_m^{(0)}(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \cos\omega(t-t_1). \quad (51)$$

In order to obtain second-order terms, we need an expression for $\gamma_m^{[1]}$. This is given by Eq. (I, 13) with $P^{(0)}$ replaced by $P^{[0]}$:

$$\gamma_m^{[1]}(t) = \frac{8\pi c}{\hbar} u \tilde{\gamma}^2 I_m \int_0^t dt_1 P^{[0]}(t_1) \sin\omega(t-t_1). \quad (52)$$

(There is no damping factor under the integral sign because the molecular oscillations are not damped internally.) We thus obtain

$$\begin{aligned} P^{[2]}(t) &= \frac{8\pi}{\hbar\beta} \omega u^2 \tilde{\gamma}^2 I_m \int_0^t dt_1 P^{[0]}(t_1) (1 - e^{-\frac{1}{2}\beta(t-t_1)}) \\ &\quad \times \cos\omega(t-t_1), \\ &\equiv (P^{[2]})_D + (P^{[2]})_\Gamma, \end{aligned} \quad (53)$$

$$\begin{aligned} Q^{[2]}(t) &= \frac{32\pi^2}{\hbar\beta} c^2 u^2 \tilde{\gamma}^2 I_m \int_0^t dt_1 P^{[0]}(t_1) (1 - e^{-\frac{1}{2}\beta(t-t_1)}) \\ &\quad \times \sin\omega(t-t_1), \\ &\equiv (Q^{[2]})_D + (Q^{[2]})_\Gamma, \end{aligned} \quad (54)$$

where an approximation based on $\beta/\omega \ll 1$ has been made, and where $(P^{[2]})_D$, $(P^{[2]})_R$ refer to the parts of $P^{[2]}$ due to P_D and P_R , respectively, according to Eq. (49) (and likewise for Q).

We can now obtain expectation values for the field strength. Up to second order in $\tilde{\gamma}$,

$$\begin{aligned} \langle \mathbf{E}(\mathbf{r}, t) \rangle = & \mathbf{E}_D(\mathbf{r}, t) \\ & + \frac{8\pi}{\beta} \omega \mathbf{u}(\mathbf{r}) |a_1 a_2| \tilde{\gamma} \sin(\omega t + \theta_m) (1 - e^{-\frac{1}{2}\beta t}) \\ & + \frac{8\pi}{\hbar \beta^2} \omega u^2 \tilde{\gamma}^2 \mathbf{E}_D(\mathbf{r}, t) (|a_2|^2 - |a_1|^2) \\ & \quad \times [\frac{1}{2}\beta t - (1 - e^{-\frac{1}{2}\beta t})], \end{aligned} \quad (55)$$

where use has been made of Eq. (I, 10). $\mathbf{E}_D(\mathbf{r}, t)$ is the driving field,⁸ given by

$$\begin{aligned} \mathbf{E}_D(\mathbf{r}, t) = & - (4\pi/\beta) \omega u \mathbf{D}(\mathbf{r}) D_0 \cos(\omega t + \theta), \\ \equiv & E_{D0} \mathbf{u}(\mathbf{r}) \cos(\omega t + \theta). \end{aligned}$$

It is to be noticed that as β approaches zero, Eq. (55) becomes identical with Eq. (I, 19), which is the expectation value of the field for the lossless case. In this comparison we identify the driving field in the lossy cavity with the zeroth-order field in the lossless cavity.

Our main interest is in the expectation value of the energy of the field. To this end we must calculate the expectation values of the squares of P and Q , respectively. We have, up to second order,

$$\langle P^2 \rangle = \langle P^{[0]2} \rangle + \langle P^{[1]2} \rangle + \langle \{P^{[0]}, P^{[1]}\} \rangle + \langle \{P^{[0]}, P^{[2]}\} \rangle, \quad (56)$$

and a similar equation for $\langle Q^2 \rangle$, where $\{A, B\} \equiv AB + BA$. Now

$$\langle P^{[0]2} \rangle = P_D^2 + \langle P_R^2 \rangle, \quad (57)$$

since, by previous reasoning,

$$\langle P_D P_R \rangle = P_D \langle P_R \rangle = 0. \quad (58)$$

We obtain $\langle P_R^2 \rangle$ from Eq. (33) by changing the lower limit of the integration to $-\infty$. We do the same for Q^2 , and obtain for the zeroth-order energy expectation value

$$\langle H_{\text{field}}^{[0]} \rangle = \frac{E_{D0}^2}{8\pi} + \frac{1}{2} \hbar \omega + \frac{\hbar \omega}{e^{\hbar \omega / kT} - 1}, \quad (59)$$

which is, of course, the sum of the energy of the driving field, the zero-point energy, and the thermal energy. The first-order term in Eq. (56) is

$$\langle \{P^{[0]}, P^{[1]}\} \rangle = 2P_D \langle P^{[1]} \rangle, \quad (60)$$

and can be obtained in the same manner as the first-order term of Eq. (55). Combining this with the first-

⁸ As our driving mechanism, we chose a dipole. There are other possible driving mechanisms for a cavity, such as a loop or an iris. It is obvious that a classical driving mechanism will produce a classical driving field in the cavity. We can, therefore, ignore the mechanism and consider the problem as specified in terms of a classical driving field, given by $\mathbf{E}_D(\mathbf{r}, t)$.

order term in Q^2 , we have, as the first-order term of the energy,

$$\langle H_{\text{field}}^{[1]} \rangle = 2\omega u \tilde{\gamma} \beta^{-1} |a_1 a_2| E_{D0} (1 - e^{-\frac{1}{2}\beta t}) \sin(\theta_m - \theta), \quad (61)$$

which corresponds to the first-order term in Eq. (I, 18), and approaches the latter for vanishing β . Equation (61) is the expression for the coherent spontaneous-emission (or absorption) energy arising from the interaction of the coherent molecular oscillations with the field, and depends, obviously, on the phase relationship between the molecular oscillation and the field oscillation.

The second-order part of $\langle P^2 \rangle$ is composed of two parts, $\langle P^{[1]2} \rangle$ and $\langle \{P^{[0]}, P^{[2]}\} \rangle$. For the former we have, from Eq. (50),

$$\begin{aligned} \langle P^{[1]2} \rangle = & \frac{\omega^2 u^2}{2c^2} \int_0^t dt_1 \int_0^t dt_2 \langle \{ \gamma^{(0)}(t_1), \gamma^{(0)}(t_2) \} \rangle \\ & \times e^{-\frac{1}{2}\beta(2t-t_1-t_2)} \sin \omega(t-t_1) \sin \omega(t-t_2). \end{aligned} \quad (62)$$

Noting, from the equation following Eq. (I, 17), that

$$\langle \{ \gamma^{(0)}(t_1), \gamma^{(0)}(t_2) \} \rangle = 2\tilde{\gamma}^2 \cos \omega(t_2 - t_1), \quad (63)$$

we obtain

$$\langle P^{[1]2} \rangle = \frac{\omega^2 u^2 \tilde{\gamma}^2}{c^2 \beta^2} (1 - 2e^{-\frac{1}{2}\beta t} + e^{-\beta t}), \quad (64)$$

where we have neglected ω^{-1} compared to β^{-1} . We also have

$$\langle \{P^{[0]}, P^{[2]}\} \rangle = 2P_D \langle (P^{[2]})_D \rangle + \langle \{P_R, (P^{[2]})_R\} \rangle. \quad (65)$$

$\langle (P^{[2]})_D \rangle$ is obtained from the last term in Eq. (55) by replacement of $E_D(t)$ by $P_D(t)$, so that

$$\begin{aligned} P_D \langle (P^{[2]})_D \rangle = & \frac{8\pi}{\hbar \beta^2} \omega u^2 \tilde{\gamma}^2 P_{D0}^2 \cos^2(\omega t + \theta) (|a_2|^2 - |a_1|^2) \\ & \times [\frac{1}{2}\beta t - (1 - e^{-\frac{1}{2}\beta t})]. \end{aligned} \quad (66)$$

Also, by methods similar to those used in deriving Eq. (33) (and using previously mentioned approximations), we obtain

$$\begin{aligned} \langle \{P_R, (P^{[2]})_R\} \rangle = & \frac{\omega^2 u^2 \tilde{\gamma}^2}{c^2 \beta^2} (|a_2|^2 - |a_1|^2) \\ & \times (1 - 2e^{-\frac{1}{2}\beta t} + e^{-\beta t}) (-\langle I_j \rangle_\omega)^{-1}. \end{aligned} \quad (67)$$

We have, thus, for the second-order part of P^2 ,

$$\frac{2\omega^2 u^2 \tilde{\gamma}^2}{c^2 \beta^2} (1 - 2e^{-\frac{1}{2}\beta t} + e^{-\beta t}) \left(|a_2|^2 + \frac{|a_2|^2 - |a_1|^2}{e^{\hbar \omega / kT} - 1} \right). \quad (68)$$

Calculating the second-order Q^2 terms in a similar manner, we obtain for the second-order energy of the

field,

$$\langle H_{\text{field}}^{[2]} \rangle = \mathcal{E}(1 - 2e^{-\frac{1}{2}\beta t} + e^{-\beta t}) \left(|a_2|^2 + \frac{|a_2|^2 - |a_1|^2}{e^{(\hbar\omega/kT)} - 1} \right) + \frac{E_{D0}^2/8\pi}{\hbar\omega} \mathcal{E}(\beta t + 2e^{-\frac{1}{2}\beta t} - 2)(|a_2|^2 - |a_1|^2), \quad (69)$$

where

$$\mathcal{E} \equiv 8\pi\omega^2 u^2 \gamma^2 / \beta^2.$$

It will prove convenient to break this up into two terms:

$$\langle H_{\text{field}}^{[2]} \rangle_{\text{spontaneous}} = \mathcal{E} |a_2|^2 (1 - 2e^{-\frac{1}{2}\beta t} + e^{-\beta t}), \quad (70)$$

and

$$\langle H_{\text{field}} \rangle_{\text{induced}} = \frac{E_{D0}^2/8\pi}{\hbar\omega} \mathcal{E}(\beta t + 2e^{-\frac{1}{2}\beta t} - 2)(|a_2|^2 - |a_1|^2) + \frac{\mathcal{E}(|a_2|^2 - |a_1|^2)(1 - 2e^{-\frac{1}{2}\beta t} + e^{-\beta t})}{e^{(\hbar\omega/kT)} - 1}. \quad (71)$$

The considerations regarding coherence and incoherence are the same as those for the lossless case, and are discussed in detail in I. According to these considerations, the second-order spontaneous emission [Eq. (70)] is incoherent, the first term of the induced emission (the part induced by the driving field) is coherent, and the second term (the part induced by the thermal field) is incoherent. The latter behaves in all respects like the spontaneous emission. The ratio of the thermally-induced emission to the spontaneous emission is

$$(e^{(\hbar\omega/kT)} - 1)^{-1} \begin{cases} \cong kT/\hbar\omega & \text{for } kT/\hbar\omega \gg 1, \\ \cong e^{-(\hbar\omega/kT)} & \text{for } \hbar\omega/kT \gg 1. \end{cases} \quad (72)$$

We see that for $\beta t \ll 1$, we have the same situation as in the lossless case, with the energy increasing from zero as the square of the time. For $\beta t \gg 1$, the spontaneous-emission energy in the cavity approaches a steady-state value; the induced-emission energy approaches a function which has a constant rate of increase as far as the coherent emission is concerned, and a steady-state value as far as the incoherent (thermally-induced) emission is concerned.

Let us consider first the case for which the temperature of the cavity is sufficiently low so that $\hbar\omega/kT \gg 1$. Then we can neglect the thermally-induced emission compared to the spontaneous emission. For a molecule in the upper state, the ratio of induced-emission energy to spontaneous-emission energy (which is also the ratio of coherent to incoherent energy) starts out initially as

$$\frac{E_{D0}^2/8\pi}{\hbar\omega}, \quad (73)$$

which is the familiar quantity expressing the (driving field) energy in the cavity in units of the energy of a

photon, and becomes, for $\beta t \gg 1$,

$$\frac{E_{D0}^2/8\pi}{\hbar\omega} \beta t. \quad (74)$$

Since the driving power is $\beta(E_{D0}^2/8\pi)$, this ratio is also equal to the number of photons sent into the cavity up to time t (ignoring transient effects). It is interesting to note that for a given driving field strength in the cavity, the steady-state value of the ratio of induced-to-spontaneous emission *increases* with increasing loss in the cavity. However, for a given driving power, the ratio of induced-to-spontaneous emission is *independent* of losses in the cavity. It is also important to note that the ratio increases with the time during which the molecule is exposed to the field. (It should be remembered that the time considered is small compared to the time in which the molecule radiates all its energy.)

For the case of $\hbar\omega/kT \ll 1$, we can neglect the spontaneous emission compared to the thermally-induced emission. As far as the time dependence and coherence properties of the thermally induced emission are concerned, they are identical to those of the spontaneous emission. In view of Eq. (72), the ratio of coherent-to-incoherent-emission energy in the cavity field, corresponding to (73) and (74), respectively, now becomes

$$\frac{E_{D0}^2/8\pi}{kT} \quad (75)$$

for $\beta t \ll 1$, and

$$\frac{E_{D0}^2/8\pi}{kT} \beta t \quad (76)$$

for $\beta t \gg 1$.

III

Although for measurement purposes and for applications (such as masers, for instance), the *field* properties are the most significant ones, the study of the molecular behavior is also of interest. Returning to the Hamiltonian of Eq. (1), we see that

$$i\hbar dH_m/dt = 4\pi cu P[H_m, \gamma_m], \quad (77)$$

so that

$$dH_m/dt = -4\pi cu P d\gamma_m/dt. \quad (78)$$

Using the expressions for $P^{[0]}$, $P^{[1]}$, and $P^{[2]}$, as well as those for $\gamma_m^{[0]}$ and $\gamma_m^{[1]}$, all of which have been derived previously, we obtain

$$\langle \dot{H}_m^{[0]} \rangle = 0, \quad (79)$$

$$\langle \dot{H}_m^{[1]} \rangle = -E_{D0} \omega u \gamma |a_1 a_2| \sin(\theta_m - \theta), \quad (80)$$

where an oscillatory term has been neglected, and

$$\langle \dot{H}_m^{[2]} \rangle = -\beta \mathcal{E} (1 - e^{-\frac{1}{2}\beta t}) \left[|a_2|^2 + \frac{|a_2|^2 - |a_1|^2}{e^{(\hbar\omega/kT)} - 1} \right] - (\omega/\hbar) u^2 \gamma^2 E_{D0}^2 t \cos^2(\omega t + \theta) (|a_2|^2 - |a_1|^2), \quad (81)$$

where the usual approximations have been made.

Let us consider first the case for which $\hbar\omega/kT \gg 1$, or very low temperature. Then the thermally-induced emission is negligible. The spontaneous (and also incoherent) power emitted by the molecule is

$$-\langle \dot{H}_m^{[2]} \rangle_{\text{spont.}} = \beta \mathcal{E} |a_2|^2 (1 - e^{-\frac{1}{2}\beta t}). \quad (82)$$

By comparing this with Eq. (70) for the spontaneous-emission energy in the field, we can see easily that

$$-\langle \dot{H}_m^{[2]} \rangle = \frac{d}{dt} \langle H_{\text{field}} \rangle_{\text{spont.}} + \beta \langle H_{\text{field}} \rangle_{\text{spont.}}, \quad (83)$$

which is equivalent to the statement that the incoherent power emitted by the molecule is equal to the rate of increase of incoherent energy in the field plus the incoherent power absorbed by the walls—a result which we would certainly expect.

We look now at the induced emission radiated by the molecule, which (for $\hbar\omega/kT \gg 1$) is given by the last term in Eq. (81). Its average value (averaged over a cycle) is

$$-\langle \dot{H}^{[2]} \rangle_{\text{induced}} = \frac{1}{2} (\omega/\hbar) u^2 \gamma^2 E_{D0}^2 t (|a_2|^2 - |a_1|^2). \quad (84)$$

By comparing this with the first term in Eq. (71), we see that we cannot make a statement such as Eq. (83) about the induced power. Thus,

$$\begin{aligned} & \frac{d}{dt} \langle H_{\text{field}}^{[2]} \rangle_{\text{induced}} + \beta \langle H_{\text{field}}^{[2]} \rangle_{\text{induced}} \\ &= \frac{\omega u^2 \gamma^2}{\beta \hbar} E_{D0}^2 (\beta t + e^{-\frac{1}{2}\beta t} - 1) (|a_2|^2 - |a_1|^2). \end{aligned} \quad (85)$$

We see that for small t it is true that the right side of Eq. (85) reduces to the right side of Eq. (84), but after the transient period the latter is only one-half the value of the former. This seemingly paradoxical situation is resolved when we realize that our driving mechanism is one of prescribed dipole moment or prescribed field. The amount of power it emits into the cavity depends on the total field in the cavity, and one can readily see that the second-order part of the power emitted by the driving mechanism is just the difference between the right sides of Eqs. (85) and (84).

The ratio of induced-to-spontaneous power, or of coherent-to-incoherent power, emitted by the molecule is (still for $\hbar\omega/kT \gg 1$)

$$\frac{1}{2} \frac{E_{D0}^2 / 8\pi}{\hbar\omega} \beta t (1 - e^{-\frac{1}{2}\beta t})^{-1}. \quad (86)$$

For small t this expression is just the number of photons in the expectation value of the cavity energy, but in the steady-state condition it increases linearly with the time and may be interpreted as one-half the total energy sent into the cavity, in units of photons, up to time t . For a temperature such that $\hbar\omega/kT \ll 1$ the same consider-

ations apply here as those for the energy in the cavity field: The ratio of coherent-to-incoherent power emitted is given by (86) with $\hbar\omega$ replaced by kT .

The result of Eq. (81) is interesting from several points of view. From a fundamental viewpoint, one should note that the spontaneous-emission power, or the spontaneous transition probability, depends on the environment of the radiator. This fact was recognized by Purcell,⁹ who gave, as the spontaneous transition probability for the case of resonance between molecule and cavity,

$$8\pi "Q" \gamma^2 / \hbar V \quad (87)$$

where V is the volume of the cavity. If we take the steady-state value (by neglecting the exponential term) of the expression for the spontaneous-emission power given in Eq. (82), average u^2 over the volume of the cavity (which gives V^{-1}),² and divide by $\hbar\omega$ (which converts power into transition probability), we obtain Purcell's formula. It should be further recognized, however, that the transition probability is not constant in time, as may be construed from Purcell's formula; and, although it approaches a steady-state value, the time taken to approach this value ($> 2/\beta$) may be considerable, depending on the "Q" of the cavity. The incoherent (thermally) induced transition probability has the same time variation as the spontaneous transition probability, as is evident from Eq. (81).

Also of fundamental interest is the fact that the coherent induced transition probability, which is the right side of Eq. (84) divided by $\hbar\omega$, is not constant at all, but increases linearly with the time. It depends on the cavity only through the strength of the driving field.

An interesting quantity is the ratio of power emitted by a molecule in the upper state to power absorbed by a molecule in the lower state, or its equivalent, the ratio of the downward-to-upward transition probability. For the case of an incoherent field in free space, this ratio has the well-known value¹⁰

$$(n+1)/n, \quad (88)$$

where n is the average number of photons per mode. For the case of a coherent field in a lossless cavity, it follows immediately from the results in I that the ratio is the same. In the present case, the situation is somewhat different.

Let us consider, first, the absence of a driving field. Then the only zero-order field in the cavity is the (incoherent) thermal field. The ratio of thermally induced to spontaneous power emitted by the molecule is the ratio of the second term to the first term in the square bracket of Eq. (81), which is, for $|a_2|^2 \sim 1$,

$$(e^{\hbar\omega/kT} - 1)^{-1}. \quad (89)$$

⁹ E. M. Purcell, Phys. Rev. **69**, 681 (1946).

¹⁰ See, for example, W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1954), third edition.

But this is, according to (40), just the number of photons in the thermal field. It follows, therefore, from Eq. (81), that the ratio of the downward- to upward-transition probability, when the induced emission is due to the thermal field, is given by (88). This, of course, was the original basis for the derivation of the Einstein transition coefficients. It can be shown that the same situation exists when the induced emission is due to any incoherent field.

The situation is different, however, in the presence of a *coherent* field. Let us consider the case for which the thermal field in the cavity is negligible. Then, the ratio of the downward- to upward-transition probabilities (averaged over a cycle) is, from Eq. (81),

$$[n + 2(\beta t)^{-1}(1 - e^{-\frac{1}{2}\beta t})]/n, \quad (90)$$

where n here, too, is the number of photons in the (driving) cavity field. For small βt this ratio approaches (88), but for $\beta t \gg 1$ it approaches unity.

The ratio of induced to spontaneous emission is also of interest in masers. Since induced emission is coherent and spontaneous emission (when the molecule is in the upper state—we are not concerned here with first-order coherent spontaneous emission) is incoherent, the induced emission is identified with signal and the spontaneous emission with noise. The ratio of the two determines the ultimate signal-to-noise ratio attainable. In previous calculations,¹¹ the ratio was considered to be n . We see now, however, from Eq. (86), that after the transient period the ratio is larger than n by a factor of $\frac{1}{2}\beta t$.

IV

Thus far we have considered only a single molecule. We can generalize our discussion quite easily to apply to a large number of molecules in the cavity. Returning to Eqs. (42) and (43), we sum the third term on the right side of both equations over all the molecules in the cavity; that is, instead of a single term we now have

$$-\frac{\omega}{c} \sum_m \int_0^t dt_1 \gamma_m(t_1) u(\mathbf{r}_m) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1) \quad (91)$$

in the expression for $\overline{P}(t)$, and

$$-4\pi c \sum_m \int_0^t dt_1 \gamma_m(t_1) u(\mathbf{r}_m) e^{-\frac{1}{2}\beta(t-t_1)} \cos\omega(t-t_1) \quad (92)$$

in the expression for $\overline{Q}(t)$. For simplicity we assume,

¹¹ M. W. P. Strandberg, Phys. Rev. **106**, 617 (1957); Shimode, Takahasi, and Townes, J. Phys. Soc. Japan **12**, 686 (1957). The author is indebted to Professor Townes for pointing out, however, that the latter reference refers to molecules which have an infinitely wide frequency response and are therefore not of the type considered in the present discussion. A separate analysis is needed for the case where there is a significant spread in molecular frequencies, and this will be given in a subsequent article. In general, a molecular-beam maser has a very narrow frequency spread, while a solid-state maser has a much more significant frequency spread.

as we did in I, that $\mathbf{u}(\mathbf{r}_m) = \mathbf{u}$ for all m . Then, by the same methods used for a single molecule, we obtain

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) = & \mathbf{E}_D(\mathbf{r}, t) + (8\pi/\beta)\omega u \tilde{\gamma} \mathbf{u}(\mathbf{r}) \\ & \times \sum_m |a_1(m)a_2(m)| (1 - e^{-\frac{1}{2}\beta t}) \sin(\omega t + \theta_m) \\ & + \mathcal{E} \mathbf{E}_D(\mathbf{r}, t) [\frac{1}{2}\beta t - (1 - e^{-\frac{1}{2}\beta t})] \\ & \times \sum_m [|a_2(m)|^2 - |a_1(m)|^2], \quad (93) \end{aligned}$$

and

$$\begin{aligned} \langle H_{\text{field}} \rangle = & \frac{E_{D0}^2}{8\pi} + \frac{1}{2}\hbar\omega + \frac{\hbar\omega}{e^{(\hbar\omega/kT)} - 1} \\ & + \frac{\omega}{\beta} 2u \tilde{\gamma} E_{D0} (1 - e^{-\frac{1}{2}\beta t}) \sum_m |a_1(m)a_2(m)| \sin(\theta_m - \theta) \\ & + \frac{E_{D0}^2/8\pi}{\hbar\omega} \mathcal{E} (\beta t + 2e^{-\frac{1}{2}\beta t} - 2) \sum_m [|a_2(m)|^2 - |a_1(m)|^2] \\ & + \mathcal{E} (1 - e^{-\frac{1}{2}\beta t})^2 \sum_m \left[|a_2(m)|^2 + \frac{|a_2(m)|^2 - |a_1(m)|^2}{e^{(\hbar\omega/kT)} - 1} \right] \\ & + \mathcal{E} \sum_{m \neq m'} |a_1(m)a_2(m)a_1(m')a_2(m')| \\ & \times (1 - e^{-\frac{1}{2}\beta t})^2 \cos(\theta_m - \theta_{m'}). \quad (94) \end{aligned}$$

We see that the expectation value of the field strength (to which only the coherent part of the field contributes) produced by all the molecules is the sum of the fields of the individual molecules. As for the energy, all the molecular terms except that last are the sums of the corresponding terms in the field energy of a single molecule. The last term gives the effect of interference (either constructive or destructive) in the coherent spontaneous emission of the molecules. The considerations pertaining to the combination of the radiation from many molecules are the same as those in the lossless case, and are discussed in detail in I.

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APPENDIX A

We show that expression (32) is equal to expression (33). Since most of the contribution to the integral of (32) comes from the terms $\omega_j \sim \omega$, we can set

$$\sum e^{i\omega_j(t_1-t_2)} = \rho(\omega) \int_0^\infty d\omega_j e^{i\omega_j(t_1-t_2)}, \quad (1A)$$

where it is important to note that ω_j assumes positive values only. Now

$$\int_0^\infty d\omega e^{i\omega\eta} = \pi\delta(\eta) + i\mathcal{P}/\eta, \quad (2A)$$

where \mathcal{P}/η is known as the principal value¹² of $1/\eta$ and has the property that

$$\int_{-a}^b d\eta f(\eta)\mathcal{P}/\eta = \lim_{\epsilon \rightarrow 0} \left[\int_{-a}^{-\epsilon} d\eta \frac{f(\eta)}{\eta} + \int_{\epsilon}^b d\eta \frac{f(\eta)}{\eta} \right].$$

The integration over the $\delta(t_1-t_2)$ term may be performed immediately, and yields (33). The integration over the $\mathcal{P}/(t_1-t_2)$ term vanishes, as shown by the following argument: Introduce a change of variables,

$$\begin{aligned} \xi &= t_1 + t_2, \\ \eta &= t_1 - t_2. \end{aligned} \quad (3A)$$

The part of the integrand multiplying \mathcal{P}/η is an even function of η . \mathcal{P}/η is an odd function of η . The limits are transformed in the manner

$$\begin{aligned} \int_0^t dt_1 \int_0^t dt_2 f(t_1, t_2) &= \frac{1}{2} \left\{ \int_0^t d\xi \int_{-\xi}^{\xi} d\eta \right. \\ &\quad \left. + \int_t^{2t} d\xi \int_{-(2t-\xi)}^{2t-\xi} d\eta \right\} \varphi(\xi, \eta), \end{aligned} \quad (4A)$$

where $f(t_1, t_2) \equiv \varphi(\xi, \eta)$. These limits are obviously symmetrical about zero for the η integration. We thus have a symmetrical integration over an odd function, which is zero.

APPENDIX B

From Eqs. (21) and (22), we have

$$\begin{aligned} [Q(t), P(t)] &= ihe^{-\beta t} \\ &\quad - 4\pi\omega U^2 \sum_j \sum_k \int_0^t dt_1 \int_0^t dt_2 \\ &\quad \times e^{-\frac{1}{2}\beta(2t-t_1-t_2)} \cos\omega(t-t_1) \sin\omega(t-t_2) \\ &\quad \times [\Gamma_j^{(0)}(t_1), \Gamma_k^{(0)}(t_2)]. \end{aligned} \quad (5A)$$

Since

$$\Gamma_j^{(0)}(t) = \bar{\Gamma} \begin{pmatrix} 0 & e^{-i\omega_j t} \\ e^{i\omega_j t} & 0 \end{pmatrix},$$

¹² See, for instance, reference 10, p. 69.

and

$$[\Gamma_j^{(0)}, \Gamma_k^{(0)}] = 0, \quad j \neq k,$$

we obtain, for the last term in Eq. (5A),

$$\begin{aligned} &- 4\pi i \omega U^2 \bar{\Gamma}^2 \int_0^t dt_1 \int_0^t dt_2 [\sin\omega(2t-t_1-t_2) \\ &\quad + \sin\omega(t_1-t_2)] e^{-\frac{1}{2}\beta(2t-t_1-t_2)} \sum_j I_j \sin\omega_j(t_1-t_2). \end{aligned} \quad (6A)$$

We make the approximation, which was used in originally obtaining Eqs. (21) and (22), of replacing I_j by its expectation value and then bringing it through the summation sign as $\langle I_j \rangle_\omega$. Noting that for purposes of evaluating (6A),

$$\begin{aligned} \sum_j \sin\omega_j \eta &= \rho(\omega) \int_0^\infty d\omega_j \sin\omega_j \eta \\ &= \rho(\omega) \mathcal{P}/\eta, \end{aligned} \quad (7A)$$

we have

$$\begin{aligned} [Q(t), P(t)] &= ihe^{-\beta t} - 2\pi i \omega U^2 \bar{\Gamma}^2 \rho(\omega) \langle I_j \rangle_\omega \\ &\quad \times \int d\xi \int d\eta e^{-\frac{1}{2}\beta(2t-\xi)} \frac{\mathcal{P}}{\eta} [\sin\omega(2t-\xi) + \sin\omega\eta], \end{aligned} \quad (8A)$$

where the limits of integration are the same as those in Eq. (4A). Carrying out the η integration first, we find that the contribution from the $\sin\omega(2t-\xi)$ term vanishes; and for the $\sin\omega\eta$ term we have

$$\int d\eta \frac{\mathcal{P}}{\eta} \sin\omega\eta = \pi \quad (9A)$$

for sufficiently large ω . Making use of Eq. (15), we obtain

$$\begin{aligned} [Q, P] &= ihe^{-\beta t} + \frac{1}{2} i h \beta \int_0^{2t} d\xi e^{-\frac{1}{2}\beta(2t-\xi)} \\ &= ihe^{-\beta t} + ih(1 - e^{-\beta t}) = ih. \end{aligned} \quad (10A)$$