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Induced and Spontaneous Emission in a Coherent Field

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The interaction between a coherently oscillating radiation field, such as is generally encountered in microwave experiments, and a number of similar atomic systems coupled to the field through their electric dipole moments is analyzed for the case of resonance between atomic system and field, with both the field and molecules treated quantum-mechanically. Expressions are derived for the expectation value of field strength and the energy, and comparison between the two makes it possible to distinguish between coherent and incoherent parts of the energy, the former having its counterpart in the expectation value of the field strength, while the latter has no such counterpart. Terms corresponding to induced and spontaneous emission are identified, and it is shown that the latter includes both coherent and incoherent components. Special situations related to masers and to the coherence of spontaneous radiation are discussed. The distinction between the behavior of correlated and uncorrelated states is examined, and it is shown that both the coherent emission from uncorrelated states and the incoherent emission from correlated states may be proportional to the square of the number of molecules. The phase of the field is predictable for the coherent emission but unpredictable for the incoherent emission.

IN radio-frequency and microwave spectroscopy, one generally deals with a coherently oscillating electromagnetic field,[1] as contrasted with optical spectroscopy, for instance, where the field is incoherent. In those cases where the behavior of the atomic system under study is the only subject of interest, and the finer aspects of the reaction of the atomic system on the field are unimportant, the field is usually treated classically, and considered to be a prescribed perturbation of the atomic system. This is the method which has been used most widely,[2] and takes proper cognizance of field coherence, since it consists of adding to the Hamiltonian of the atomic system a sinusoidally time-varying potential. In the cases, however, where the details of behavior of the field itself, as affected by the atomic system, are of interest, the field must be included as part of the entire system under study, and treated quantum-mechanically. This has been done, to greater or lesser extent, with respect to such problems as noise in masers,[3–5] and the coherence of spontaneous emission[6]; but here, however, there has not been complete recognition of the coherence properties of the field, since the formalism used describes the field either by means of a smooth intensity function of the frequency[3,4,6] or as a collection of independent photons.[5]

The purpose of the present article is to examine some important aspects of the behavior of a coherently oscillating radiation field when it is in resonance with a group of similar atomic systems, no coupling existing between the systems themselves other than through the field. As a specific illustration of such a system, we may think of an ammonia beam maser.[7] The field is the radiation field in the microwave cavity, and the atomic systems are the ammonia molecules. The molecular resonance is that of the inversion spectrum, and the coupling to the field is due to the electric dipole moment of the molecule. However, the analysis is in no way specialized by any references to molecular structure, and is presented only in terms of an electric dipole moment.

The questions to be examined are those relating to induced emission, spontaneous emission, coherence, and correlation effects between molecules. In part I, expectation values for the field strength and field energy are derived, and the physical origin of the terms occurring in these expectation values is discussed. In part II, we deal with the question of coherence. In part III, situations associated with masers and with spontaneous radiation from excited groups of molecules are analyzed. In part IV, the distinction between correlated and uncorrelated states is studied.

I.

We shall consider an idealized situation so that we may deal only with the bare essentials of the problem and concentrate on the principles involved, rather than complicate it with details which do not affect the results significantly but make the computation more difficult. The atomic system, to which we refer henceforth as "molecule," is assumed to have only two internal energy states with eigenvalues E_1 and E_2, $E_2 > E_1$. The motion of the center of mass of the molecule is assumed to be classical. The radiation field is contained in a resonant cavity which is lossless. We consider only one mode of the cavity, for which we assume a condition of exact resonance with the molecule, namely, $\omega = (E_2 - E_1)/\hbar$.

The field is described in the usual manner by

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

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[1] By coherent oscillation we mean a sinusoidal oscillation with well-defined phase.

[2] See, for instance, N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956).

[3] M. W. P. Strandberg, Phys. Rev. **106**, 617 (1957).

[4] R. V. Pound, Ann. Phys. **1**, 24 (1957).

[5] Shimode, Takahasi, and Townes, J. Phys. Soc. Japan **12**, 686 (1957).

[6] R. H. Dicke, Phys. Rev. **93**, 99 (1954).

[7] Gordon, Zeiger, and Townes, Phys. Rev. **99**, 1264 (1955).

where Q and P are the quantum-mechanical variables of the radiation field, obeying the commutation rule $[Q,P]=i\hbar$. The initial state of the field is described by the wave function[8] (in the P representation)

$$\varphi(P,0)=\left(\frac{4c^2}{\hbar\omega}\right)^{\frac{1}{4}}\exp\left[-\frac{4\pi c^2}{\hbar\omega}\left(\tfrac{1}{2}P^2-\frac{E_0}{4\pi c}P\right)\right]. \tag{2}$$

It is the choice of state of the field which determines whether one is describing a coherently oscillating field[9] (in the classical sense), and this is an essential aspect of the present analysis. That such is the case for the state described by Eq. (2) is shown in detail in reference 8. The molecules are described initially by the state[10] (in the energy representation of the free molecule)

$$\psi=\prod_{m=1}^{N}\left[a_1(m)\varphi_1(m)+a_2(m)\varphi_2(m)\right], \tag{3}$$

where $\varphi_1(m)$ and $\varphi_2(m)$ are the two energy states of the mth free molecule, and

$$|a_1(m)|^2+|a_2(m)|^2=1. \tag{3a}$$

The electric dipole moment of the mth molecule is specified by $\boldsymbol{\gamma}_m$ which, in the absence of the electromagnetic field, has no diagonal matrix elements (this means that the molecule has no permanent dipole moment) but only off-diagonal elements. The Hamiltonian for the combined system of molecules and field is

$$H=\sum_{m=1}^{N}H_m-\sum_m\boldsymbol{\gamma}_m\cdot\mathbf{E}(\mathbf{r}_m)+2\pi c^2P^2+\frac{\omega^2}{8\pi c^2}Q^2, \tag{4}$$

where H_m is the Hamiltonian and $\mathbf{r}_m$ the position of the mth molecule. H_m refers only to the internal energy of the molecule, and not to the kinetic energy of the center of mass, which we ignore. The Heisenberg equations of motion for the field variables are

$$\dot{P}=-(\omega^2/4\pi c^2)Q,$$
$$\dot{Q}=4\pi c^2P+4\pi c\textstyle\sum_m\boldsymbol{\gamma}_m\cdot\mathbf{u}(\mathbf{r}_m).$$

These may also be expressed as integral equations which contain the initial values of P and Q explicitly:

$$\begin{aligned}Q(t)&=Q^{(0)}(t)+4\pi c\sum_m\int_0^t dt_1\,\boldsymbol{\gamma}_m(t_1)\\&\qquad\cdot\mathbf{u}(\mathbf{r}_m)\cos\omega(t-t_1),\\P(t)&=P^{(0)}(t)-\frac{\omega}{c}\sum_m\int_0^t dt_1\,\boldsymbol{\gamma}_m(t_1)\\&\qquad\cdot\mathbf{u}(\mathbf{r}_m)\sin\omega(t-t_1),\end{aligned} \tag{5}$$

where

$$P^{(0)}(t)=P(0)\cos\omega t-(\omega/4\pi c^2)Q(0)\sin\omega t,$$
$$Q^{(0)}(t)=Q(0)\cos\omega t+(4\pi c^2/\omega)P(0)\sin\omega t.$$

We see that $P^{(0)}(t)$ and $Q^{(0)}(t)$ describe the behavior of the field in the absence of an interaction with the molecules. For future reference, we note the expectation values of $P^{(0)}(t)$, $P^{(0)2}(t)$, as well as $Q^{(0)}(t)$ and $Q^{(0)2}(t)$, which are obtained from Eq. (2)[8]:

$$\begin{aligned}\langle P^{(0)}\rangle&=-(E_0/4\pi c)\sin\omega t,\\\langle P^{(0)2}\rangle&=\langle P^{(0)}\rangle^2+\hbar\omega/8\pi c^2,\\\langle Q^{(0)}\rangle&=(E_0c/\omega)\cos\omega t,\\\langle Q^{(0)2}\rangle&=\langle Q^{(0)}\rangle^2+2\pi c^2\hbar/\omega.\end{aligned} \tag{6}$$

The summations on the right side of Eqs. (5) are due to the coupling between the molecules and field. We assume that the dipole moment of the molecule is sufficiently small so that it may be regarded as a small quantity of first order, and expand our dynamical variables in powers of the coupling constant (which is contained in $\boldsymbol{\gamma}$).

Setting

$$P(t)=P^{(0)}(t)+P^{(1)}(t)+P^{(2)}(t)+\cdots,$$

and using similar expansions for $Q(t)$ and $\boldsymbol{\gamma}(t)$, we have, from Eq. (5),

$$Q^{(n)}(t)=4\pi c\sum_m\int_0^t dt_1\,\boldsymbol{\gamma}_m{}^{(n-1)}(t_1)\cdot\mathbf{u}(\mathbf{r}_m)\cos\omega(t-t_1), \tag{7}$$

$$P^{(n)}(t)=-\frac{\omega}{c}\sum_m\int_0^t dt_1\,\boldsymbol{\gamma}_m{}^{(n-1)}(t_1)\cdot\mathbf{u}(\mathbf{r}_m)\sin\omega(t-t_1). \tag{8}$$

We make a further simplification for the purpose of disencumbering our analysis of complicating features which have no bearing on the essence of the problem. We will consider $\mathbf{u}(\mathbf{r})$ to be constant in the region of the cavity where the molecules are located. The component of $\boldsymbol{\gamma}_m$ along the direction of $\mathbf{u}$ will be designated by γ_m, with matrix elements for $\gamma_m{}^{(0)}(0)$ given by $\gamma_{m12}=\gamma_{m21}{}^*=\tilde{\gamma}_m$. We chose the phases of $\varphi_1(m)$ and $\varphi_2(m)$, respectively, so as to make $\tilde{\gamma}_m$ real.

The expectation values of $Q^{(1)}(t)$ and $P^{(1)}(t)$ may be calculated immediately. Since the formal equation of motion for $\gamma_m(t)$ is

$$i\hbar\dot{\gamma}_m(t)=[\gamma_m(t),H]=[\gamma_m(t),H_m(t)], \tag{9}$$

the off-diagonal terms of $\gamma^{(0)}(t)$ have the time dependence of $\exp(\mp i\omega t)$, respectively, so that

$$\begin{aligned}\langle\gamma_m{}^{(0)}(t)\rangle&=a_1{}^*(m)a_2(m)\tilde{\gamma}e^{-i\omega t}+a_1(m)a_2{}^*(m)\tilde{\gamma}e^{i\omega t}\\&=2|a_1(m)a_2(m)|\,\tilde{\gamma}\cos(\omega t+\theta_m),\end{aligned} \tag{10}$$

where θ_m is the difference in phase between $a_2(m)$ and $a_1(m)$. We thus obtain

$$\begin{aligned}\langle P^{(1)}(t)\rangle&\cong-(\omega/c)\textstyle\sum_m|a_1(m)a_2(m)|\,\tilde{\gamma}ut\\&\qquad\times\sin(\omega t+\theta_m),\end{aligned} \tag{11}$$

$$\langle Q^{(1)}(t)\rangle\cong4\pi c\textstyle\sum_m|a_1(m)a_2(m)|\,\tilde{\gamma}ut\cos(\omega t+\theta_m),$$

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[8] I. R. Senitzky, Phys. Rev. **95**, 904 (1954); **98**, 875 (1955).

[9] A quantum-mechanical system may also be described by a mixture of states, in which the phase of the superposition constants is random. The choice of a pure state rather than a mixture is itself a step in the direction of coherence.

[10] This is an uncorrelated state. Correlated states will be discussed in part IV.

where, for simplicity, we have dropped terms in ω^{-1} compared to t. The individual terms of the summation are the contributions of the individual molecules to the field. Although the expectation values of P and Q are, strictly speaking, quantum-mechanical averages over an ensemble of similar systems, we may regard them, in this instance, as an average over similar molecules, since only molecular parameters are involved on the right side of Eqs. (11). We have here, in the first order, spontaneous emission (referring now only to the expectation value of the field; the energy will be discussed later), each molecule acting (on the average) as a (classical) oscillator of well defined phase, θ_m, and amplitude which is proportional to $|a_1(m)a_2(m)|$. We see that this amplitude is a maximum when $|a_1(m)| = |a_2(m)| = 2^{-\frac{1}{2}}$ (that is, when the molecule is in a state which is a superposition of equal amounts of both energy states), and that the amplitude vanishes when the molecule is completely in either one of the energy states.

In order to evaluate $P^{(2)}$, we need an expression for $\gamma_m^{(1)}$ in terms of the zero-order (uncoupled) operators. From Eq. (9) we have

$$i\hbar\dot{\gamma}_m^{(1)}(t)=[\gamma_m^{(1)}(t),H_m^{(0)}]+[\gamma_m^{(0)}(t),H_m^{(1)}(t)]. \tag{12}$$

An expression for $H_m^{(1)}$ can be obtained immediately from the equation of motion for H_m,

$$i\hbar\dot{H}_m(t)=-E[H_m,\gamma_m],$$

$$H_m(t)=H_m(0)+\frac{4\pi c}{i\hbar}u\int_0^t dt_1[H_m(t_1),\gamma_m(t_1)]P(t_1),$$

$$H_m^{(1)}(t)=\frac{4\pi c}{i\hbar}u\int_0^t dt_1[H_m^{(0)},\gamma_m^{(0)}(t_1)]P^{(0)}(t_1).$$

We see that $H_m^{(1)}$ contains only off-diagonal matrix elements (with reference to the molecule), so that the second term on the right side of Eq. (12) contains only diagonal elements. On the other hand, the first term on the right side of Eq. (12) contains only off-diagonal elements, so that the equation for $\gamma_{m12}^{(1)}$ is the same as for $\gamma_{m12}^{(0)}$, namely $\dot{\gamma}_{m12}^{(1)}=-i\omega\gamma_{m12}^{(1)}$. Since, however, $\gamma_m^{(1)}(0)=0$, we must have $\gamma_{m12}^{(1)}=0$. Similarly, $\gamma_{m21}^{(1)}=0$. The first term on the right side of Eq. (12) therefore vanishes. Noting that

$$[\gamma_m^{(0)}(t),[\gamma_m^{(0)}(t_1),H_m^{(0)}]]=2\hbar\omega\tilde{\gamma}^2 I_m\cos\omega(t-t_1),$$

where $I_m\equiv\begin{pmatrix}-1 & 0\\ 0 & 1\end{pmatrix}$ for the mth molecule and the unit matrix for the other molecules, we obtain

$$\gamma_m^{(1)}(t)=\frac{8\pi c}{\hbar}u\tilde{\gamma}^2 I_m\int_0^t dt_1\,P^{(0)}(t_1)\sin\omega(t-t_1) \tag{13}$$

and

$$P^{(2)}(t)=\frac{8\pi\omega}{\hbar}u^2\tilde{\gamma}^2\int_0^t dt_1\,P^{(0)}(t_1)\times\left[\tfrac{1}{2}(t-t_1)\cos\omega(t-t_1)-\frac{1}{2\omega}\sin\omega(t-t_1)\right]\sum_m I_m. \tag{14}$$

For $t\gg\omega^{-1}$, this becomes

$$P^{(2)}(t)\cong\frac{\mathcal{E}_0(t)}{2\hbar\omega}P^{(0)}(t)\sum_m I_m, \tag{15}$$

where $\mathcal{E}_0(t)\equiv 2\pi\omega^2u^2\tilde{\gamma}^2t^2$. Similarly, we have

$$Q^{(2)}(t)\cong\frac{\mathcal{E}_0(t)}{2\hbar\omega}Q^{(0)}(t)\sum_m I_m.$$

We recognize immediately that this is induced radiation proportional to the zero-order field. For the expectation values, we have

$$\langle\gamma_m^{(1)}(t)\rangle\cong E_0u\hbar^{-1}\tilde{\gamma}^2t[|a_2(m)|^2-|a_1(m)|^2]\cos\omega t,$$

$$\langle P^{(2)}(t)\rangle\cong-\frac{E_0}{8\pi c}\frac{\mathcal{E}_0(t)}{\hbar\omega}\times\sin\omega t\sum_m[|a_2(m)|^2-|a_1(m)|^2]. \tag{16}$$

We see that (on the average) the induced radiation of each molecule is in phase (emission) or 180° out of phase (absorption) with respect to the inducing field, depending upon whether the molecule is mostly in the upper energy state or lower energy state.

It is worth noting that if $E_0=0$, that is, if there is no oscillating field initially in the cavity, and if the molecule enters the cavity in either the upper or lower energy state (but not in a superposition of both), then the expectation value of the field remains zero. We have shown this to be true up to second order, but it can be shown by a general theorem to be true for all orders, as follows: consider the Hamiltonian of Eq. (4). It is invariant under a unitary transformation which changes the sign both of $\mathbf{E}$ and $\boldsymbol{\gamma}$. A state of the system for which each molecule is in a definite energy state must remain unchanged except for multiplication by a phase factor (which has no physical significance). The expectation value of the field should be invariant under the above transformation. It must therefore be zero.

We shall now calculate the expectation value for the energy of the field. This will give us information not only about the energy itself, but also about the coherence of the field. We have

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

Our calculation will include terms only up to second order. Designating the individual terms in the sum of Eq. (8) by $P_m^{(n)}$, we can write

$$P^2=P^{(0)2}+2P^{(0)}P^{(1)}+2P^{(0)}P^{(2)}+\textstyle\sum_{m,m'}P_m^{(1)}P_{m'}^{(1)}. \tag{17}$$

We need not symmetrize our products since $P^{(0)}$, $P^{(1)}$, and $P^{(2)}$ all commute.[11] $P^{(0)2}$ occurs in both the first and third terms on the right side of Eq. (17) [see Eq. (15)], and its expectation value is given in Eq. (6). The expectation value of $P^{(0)}P^{(1)}$ is just the product of the expectation values of $P^{(0)}$ and $P^{(1)}$, which are given by Eqs. (11) and (6). To obtain the expectation value of the last term in Eq. (17), we must exercise a little care. For $m\neq m'$, $\langle P_m{}^{(1)}P_{m'}{}^{(1)}\rangle=\langle P_m{}^{(1)}\rangle\langle P_{m'}{}^{(1)}\rangle$. For $m=m'$, however, we must go back to Eq. (9) and note that

$$\begin{aligned}\langle\gamma_m{}^{(0)}(t_1)\gamma_m{}^{(0)}(t_2)\rangle\\ &=\tilde{\gamma}^2[|a_1(m)|^2e^{i\omega(t_2-t_1)}+|a_2(m)|^2e^{-i\omega(t_2-t_1)}],\end{aligned}$$

so that

$$\begin{aligned}\langle P_m{}^{(1)2}\rangle&=\frac{\omega^2}{c^2}u^2\tilde{\gamma}^2\left|\int_0^t dt_1e^{i\omega t_1}\sin\omega(t-t_1)\right|^2\\ &=\frac{\omega^2}{c^2}u^2\tilde{\gamma}^2\frac{1}{4}\left(t^2-\frac{t}{\omega}\sin\omega t\cos\omega t+\frac{1}{\omega^2}\sin^2\omega t\right)\\ &\cong\frac{\mathcal{E}_0(t)}{8\pi c^2},\quad t\gg\omega^{-1}.\end{aligned}$$

Combining the above results, we obtain for the part of the energy due to the electric field

$$\begin{aligned}&2\pi c^2\langle P^2\rangle\\ &\quad=\frac{E_0}{8\pi}\sin^2\omega t\left\{1+\frac{\mathcal{E}_0(t)}{\hbar\omega}\sum_m[|a_2(m)|^2-|a_1(m)|^2]\right\}\\ &\quad\quad+E_0\omega\tilde{\gamma}ut\sum_m|a_1(m)a_2(m)|[\cos\theta_m\\ &\quad\quad-\cos(2\omega t+\theta_m)]+\tfrac{1}{2}\mathcal{E}_0(t)\{\sum_m|a_2(m)|^2\\ &\quad\quad+\sum_{m\neq m'}|a_1(m)a_2(m)||a_1(m')a_2(m')|[\cos(\theta_m-\theta_{m'})\\ &\quad\quad\quad-\cos(2\omega t+\theta_m+\theta_{m'})]\}+\tfrac{1}{4}\hbar\omega.\end{aligned}$$

Calculating $\langle Q^2\rangle$ in the same manner as we did $\langle P^2\rangle$, and adding the magnetic to the electric energy, we find the double-frequency terms cancel, and we have

$$\begin{aligned}\langle H_{\text{field}}\rangle=&\frac{E_0^2}{8\pi}+\frac{E_0^2}{8\pi}\frac{\mathcal{E}_0(t)}{\hbar\omega}\sum_m[|a_2(m)|^2-|a_1(m)|^2]\\ &+E_0\omega\tilde{\gamma}ut\sum_m|a_1(m)a_2(m)|\cos\theta_m\\ &+\mathcal{E}_0(t)\sum_m|a_2(m)|^2\\ &+\mathcal{E}_0(t)\sum_{m\neq m'}|a_1(m)a_2(m)||a_1(m')a_2(m')|\\ &\times\cos(\theta_m-\theta_{m'})+\tfrac{1}{2}\hbar\omega.\qquad(18)\end{aligned}$$

The physical meaning of the various terms is as follows: The first term is, of course, the energy of the initial field. The second term is induced radiation, and is, in the case of each molecule, emission or absorption, depending on whether the molecule is mainly in the higher energy state or lower energy state. The third term is the interaction between the (initial) oscillation of the molecule and the field which exists in the cavity. The fourth and fifth terms are spontaneous emission, and the last term is the zero-point energy of the field. Much more can be said about these terms, especially for special initial situations which are of interest, and we will come back to a discussion of these terms after we have discussed the question of coherence.

We write down, for completeness, the entire expression for the expectation value of the electric field strength (up to second order), by combining Eqs. (1), (11), and (16):

$$\begin{aligned}\langle E\rangle=&E_0u\sin\omega t\\ &+E_0u\frac{\mathcal{E}_0(t)}{2\hbar\omega}\sum_m[|a_2(m)|^2-|a_1(m)|^2]\sin\omega t\\ &+4\pi\omega u^2t\tilde{\gamma}\sum_m|a_1(m)a_2(m)|\sin(\omega t+\theta_m).\qquad(19)\end{aligned}$$

As has already been indicated, the first term is the initial cavity field, the second is an induced field, and the third is a spontaneously emitted field.

II.

By coherent oscillation, as mentioned previously, we mean a sinusoidal oscillation with a well-defined phase. By coherent radiation from a molecule, we mean radiation for which the field oscillates coherently, with the phase being determined by the state of the molecule or by the (coherent) field to which the molecule is coupled. If the state of the molecule and/or coupled field do not determine the phase of the radiation, then the radiation is incoherent and the phase is a random variable. Obviously, only coherent radiation will contribute to an average of the radiation field over many similar systems, but the incoherent radiation *will* show up in an average of the square of the field or (essentially) the energy. Thus, those terms which appear in the expectation value of the field energy but have no counterpart in the expectation value of the field strength denote the incoherent radiation. The other terms denote the coherent radiation. Another way of saying this is that the *dispersion* in field $(\langle E^2\rangle-\langle E\rangle^2)$ is a measure of the incoherence. However, it is more convenient to deal with the energy, and we will examine the terms in Eq. (18) from the point of view of coherence.

We see immediately that the first term in the expression for the energy corresponds to the square of the first term in $\langle E\rangle$($\sin^2\omega t$ combines with $\cos^2\omega t$ from the magnetic part of the energy), and is just the classical

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[11] $P^{(0)}$ contains only field variables and $P^{(1)}$ contains only molecular variables.

expression for the energy of the initial field, or the classical energy of the field in absence of the molecules. It is, of course, coherent energy. The second and third terms in $\langle H_{\text{field}}\rangle$ similarly correspond to terms in the square of $\langle E\rangle$, namely, the cross-products, and therefore also denote coherent energy. So far, the only term in the square of $\langle E\rangle$ (up to second order) which has not been accounted for in Eq. (18) is the square of the last term in $\langle E\rangle$, for which the corresponding term in the energy would be

$$\begin{aligned}\mathcal{E}_0(t) &\sum_{m,m'} |a_1(m)a_2(m)||a_1(m')a_2(m')| \cos(\theta_m-\theta_{m'})\\ &=\mathcal{E}_0(t)\{\sum_{m\neq m'} |a_1(m)a_2(m)||a_1(m')a_2(m')| \cos(\theta_m-\theta_{m'})\\ &\qquad+\sum_m [|a_2(m)|^2-|a_2(m)|^4]\},\qquad(20)\end{aligned}$$

where use has been made of Eq. (3a). This represents part of the fourth term and the entire fifth term in $\langle H_{\text{field}}\rangle$, and thus is the coherent spontaneous emission energy. The remainder of the fourth term is

$$\mathcal{E}_0(t) \textstyle\sum_m |a_2(m)|^4,\qquad(21)$$

and represents the *incoherent* spontaneous emission energy. The last term in Eq. (18) which, as has been noted previously, is the zero-point energy of the field, is independent of the molecules, and, according to our definition, would be classified as incoherent energy. However, this is energy which cannot be withdrawn from the field, does not consist of oscillation or fluctuation in time, cannot be detected in a classical-type experiment, and will be neglected for purposes of the present article. In sum, the incoherent part of the emitted radiation is given by (21); the remainder of the radiation energy, whether induced or spontaneous, is coherent, the former having its phase determined by the inducing field, and the latter having its phase determined by the initial state of the molecule.

We can gain further insight into the spontaneous emission, which will sharpen the differentiation between the coherent and incoherent parts, and make our definition more flexible. We note that the first-order part of $\langle E\rangle$, and the coherent part of the spontaneous emission energy, (which corresponds to the square of the former), are the field and energy, respectively, of a classical radiation field coupled to a classically oscillating dipole moment, $\langle\gamma(t)\rangle$. The incoherent part of the spontaneous emission energy, on the other hand, is due to the quantum-mechanical properties of the molecule and field. Thus, the coherent spontaneous emission may be regarded as of classical origin, while the incoherent spontaneous emission is of quantum-mechanical origin.

It may happen that we do not know, or cannot control, the initial states of the molecules, and are faced with a random distribution of initial states. This means that the "coherent" radiation of each molecule has random phase and should more properly be called "incoherent." This is a classical type of incoherence, such as would result if we had an assembly of classical oscillators with random initial phase. As long as we are aware of this possibility and take it into account when necessary, we will not be misled by our definitions, and will continue to use them as defined previously.

III.

We return now to Eq. (18) to examine certain situations which are of interest. Suppose, for simplicity, that $|a_1(m)a_2(m)|=|a_1(m')a_2(m')|$; that is, the amplitudes of oscillation of all the molecules are equal. Now consider the case where half the molecules have one phase ($\theta_m=\theta$, $m=1,\cdots,\frac{1}{2}N$) and the other half has opposite phase ($\theta_m=\theta+\pi$, $m=\frac{1}{2}N+1,\cdots,N$). Then the coherent spontaneous radiation [Eq. (20) for the energy or last term of Eq. (19) for the field strength] vanishes. On the other hand, if M molecules have one phase and the remainder have opposite phase, then the coherent spontaneous emission energy is proportional to $(2M-N)^2$, and, of course, for the case in which all molecules are in phase, the energy is proportional to N^2. This last situation, for the case where $|a_1(m)a_2(m)|=\frac{1}{2}$, gives the maximum possible coherent emission. It should be noted, however, that the incoherent spontaneous emission is entirely independent of the phase of the molecule, and even in the case where the coherent spontaneous emission vanishes, there is incoherent spontaneous emission, the energy being proportional to the total number of molecules, as is evident from Eq. (21).

A situation which is of interest in connection with spontaneous emission is one in which the molecules are subjected to a strong field for a short time, after which the field is removed and the molecules are allowed to radiate by themselves. To observe the effect of the field on the molecules, we need consider only a single molecule, since the interaction between molecules does not enter in the lowest order necessary to study this effect. For simplicity, we assume that the molecule is in the ground state. From Eq. (13) we obtain

$$\begin{aligned}\langle\gamma^{(1)}\rangle&=(E_0u/\hbar\omega)\tilde{\gamma}^2(\omega t\cos\omega t-\sin\omega t),\\ &\cong(E_0u/\hbar)\tilde{\gamma}^2t\cos\omega t,\quad t\gg\omega^{-1}.\end{aligned}$$

The expectation value of the energy absorbed by the molecule from the field is just the negative of the second term in Eq. (18), so that

$$\langle H_m\rangle=E_1+\frac{E_0^2}{8\pi}\frac{\mathcal{E}_0(t)}{\hbar\omega}.$$

When the field is removed, at time τ, say, we have a new set of initial conditions for the molecule, which is now in the state $a_{1\tau}\varphi_1+a_{2\tau}\varphi_2$. Since the expectation value of the dipole moment and energy of the molecule suffer no discontinuity at $t=\tau$, we have

$$2|a_{1\tau}a_{2\tau}|\tilde{\gamma}\cos(\omega\tau+\theta_\tau)=(E_0u/\hbar)\tilde{\gamma}^2\tau\cos\omega\tau,$$

and

$$|a_{1\tau}|^2E_1+|a_{2\tau}|^2E_2=E_1+\frac{E_0^2}{8\pi}\frac{\mathcal{E}_0(\tau)}{\hbar\omega}.$$

Together with the equation

$$|a_{1\tau}|^2+|a_{2\tau}|^2=1,$$

we have three equations for the determination of $|a_{1\tau}|$, $|a_{2\tau}|$, and θ_τ. The results are[12]

$$\begin{aligned}|a_{2\tau}|&=E_0u\tilde{\gamma}\tau/2\hbar,\quad \theta_\tau=0,\\ |a_{1\tau}|&=1-E_0^2\mathcal{E}_0(\tau)/16\pi(\hbar\omega)^2.\end{aligned}\tag{22}$$

We see that at time τ we have an induced oscillating dipole moment with a well-defined phase determined by the phase of the inducing field. After the field is removed, this will act as an "initial" dipole moment which will give rise to "spontaneous" coherent emission. It can be shown easily that no matter what the initial state of the molecule is at the start of the pulse (except for the case $a_1=a_2$), it will have an induced dipole moment at the end of the pulse (in addition to the initial dipole moment, and a small random moment due to spontaneous incoherent emission during the pulse), which oscillates with a phase determined by the inducing field and which is the same for molecules having the same sign of $(|a_2|^2-|a_1|^2)$. We conclude, therefore, that molecules excited by a coherent rf field will subsequently emit coherent spontaneous radiation, the phase of which is determined by the exciting field.

Another case which is of interest is the one in which the molecules are all originally in the upper state. This is the situation in the ammonia beam maser, where there is selection of states before the molecules enter the cavity. In this case the molecules emit induced energy given by

$$N\frac{E_0^2}{8\pi}\frac{\mathcal{E}_0(t)}{\hbar\omega},\tag{23}$$

and incoherent spontaneously emitted energy given by $N\mathcal{E}_0(t)$. The ratio of induced, coherent, energy to spontaneously emitted, incoherent, energy is

$$\frac{(E_0^2/8\pi)}{\hbar\omega},\tag{24}$$

namely, the expectation value of the energy in the cavity (ignoring the zero-point energy) in units of energy of a photon. For the case in which the initial state of the field is an energy state, it is well known that the ratio of induced emission to spontaneous emission is equal to the number of photons in the initial state.[13] Equation (24) is the corresponding statement for our classical type state which cannot be described in terms of an integral number of photons.

Expression (23) for the induced emission does not tell the whole story, however. We see from Eq. (5) or Eq. (7) that the field radiated by the molecule is due to the oscillating dipole moment of the molecule (as is, of course, physically evident). The only difference, essentially, between induced emission and coherent spontaneous emission is the manner in which the oscillations of the dipole moment are produced. For coherent spontaneous emission, we assume implicitly a given oscillation of the dipole moment in the initial conditions. In induced emission, the oscillations of the dipole moment are produced by the field acting on the molecules. But it is evident from the formalism, as well as from physical considerations, that the radiated field should not depend on *how* the molecular oscillations are produced, but merely on the phase and amplitude of these oscillations. This is indeed the case.

First, let us look at Eq. (19) and compare the parts of the field due to induced and spontaneous emission, respectively. The field due to (coherent) spontaneous emission increases linearly with time. This is certainly to be expected for the case of a lossless cavity, an oscillating dipole of constant amplitude, and exact resonance. The induced emission field amplitude, however, increases as the square of the time. The explanation is that the amplitude of the oscillating dipole induced by the field does itself increase linearly with the time. In fact, a comparison of the two terms shows that the induced radiation field of a molecule at time t is the same as the coherent spontaneous radiation field of the molecule if it were, during the time $0-t$, in the state described by a_1', a_2' and θ', such that $\theta'=\pi$ and

$$|a_1'a_2'|=\tfrac{1}{4}E_0u\tilde{\gamma}t(|a_2|^2-|a_1|^2).\tag{25}$$

This is consistent with the result obtained in Eq. (22).

We now come to a more important matter in the comparison of induced and coherent spontaneous emission. The induced emission of Eq. (23) is proportional to the number of molecules, N. We have seen that induced radiation comes from the induced oscillation of the molecules which is of the same phase for all molecules. In the case of coherent spontaneous emission, on the other hand, when all molecules oscillate in phase, the energy radiated is proportional to N^2. There seems to be thus, at first glance, a significant difference in the nature of the radiation. This difference is only formal, however. In the expression for the energy [Eq. (18)] we have kept only terms up to second order in $\tilde{\gamma}$. In the case of induced oscillation of the molecule, $|a_1'a_2'|$ in Eq. (25) is proportional to $\tilde{\gamma}$, so that if we wanted to regard the induced emission in the same manner as the coherent spontaneous emission, we would have a term of fourth order in $\tilde{\gamma}$. As a matter of fact, we can see immediately that the square of $P^{(2)}$ [Eq. (14)] gives

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[12] The reason we can determine the state of a system from the expectation values of two operators is that we are dealing here with a very simple system, one which has only two energy eigenvalues.

[13] The transition probability from a state of n photons to a state of $n+1$ photons is proportional to $n+1$, which may be regarded as a sum of two terms, the term proportional to n being induced emission and the remaining term being spontaneous emission.

us an induced-emission term in the energy proportional to N^2.

The present analysis, which is based on perturbation theory, cannot be used without significant modification to study quantitatively the case where higher order terms are larger than lower order terms. It is apparent, however, in a qualitative manner, that as the induced oscillations of the molecule become large (our perturbation theory is correct when these oscillations are small, and, to the first order, these oscillations increase linearly with the time) the term proportional to N^2, referred to above, will exceed the term proportional to N. In the ammonia-beam maser the molecules spend a sufficiently long time in the cavity for this to be true.

For the case in which there is no initial field in the cavity, and the molecules come into the cavity in the upper state, Eq. (18) tells us how the incoherent radiation energy will build up. If a molecule spends a time τ in the cavity, and M molecules per second enter the cavity, then the rate at which energy is given up to the cavity is $M\mathcal{E}_0(\tau)$. For ammonia molecules, perturbation theory is entirely adequate in this case, since the time a molecule spends in the cavity is very much smaller than the lifetime of the upper state.[7]

IV.

The initial molecular states we have considered so far are products of one-molecule states, and are therefore uncorrelated states; that is, measurements of the state of each molecule are, in principle, independent of one another. There is, however, another class of states, namely, correlated states. These are linear combinations of products of one-molecule states.[14]

Before we discuss our N-molecule system, it will be instructive to consider a system of two molecules, in order to examine the significance of correlated states from the present point of view. Let us consider the two possible correlated initial states[15]

$$\psi_s = 2^{-\frac{1}{2}}[\varphi_1(1)\varphi_2(2)+\varphi_2(1)\varphi_1(2)], \tag{26}$$

and

$$\psi_a = 2^{-\frac{1}{2}}[\varphi_1(1)\varphi_2(2)-\varphi_2(1)\varphi_1(2)]. \tag{27}$$

Any other correlated state may be considered as a superposition of these two states and the states $\varphi_1(1)\varphi_1(2)$ and $\varphi_2(1)\varphi_2(2)$.

We calculate the expectation value of the electric field strength and of the energy for ψ_s and ψ_a, just as we have done for the uncorrelated states. Noting that

$$\langle \gamma_1^{(0)}+\gamma_2^{(0)}\rangle_{a,s}=0,$$

and that

$$\langle I_1+I_2\rangle_{a,s}=0,$$

we see, from Eqs. (8) and (15), respectively, that

$$\langle P^{(1)}\rangle_{a,s}=\langle P^{(2)}\rangle_{a,s}=0,$$

that is, the expectation value of the field strength vanishes. As for the energy, we obtain, by methods already used, the results

$$\sum_{m,m'=1}^{2}\langle P_m^{(1)}P_{m'}^{(1)}\rangle_s=\frac{\mathcal{E}_0(t)}{2\pi c^2},$$

$$\sum_{m,m'=1}^{2}\langle P_m^{(1)}P_{m'}^{(1)}\rangle_a=0.$$

The expectation value of the other terms in Eq. (17), except that of $(P^{(0)})^2$, vanishes for both ψ_a and ψ_s. We thus obtain, for the field energy due to the molecules,

$$\langle H_{\text{field}}\rangle_s=2\mathcal{E}_0(t),\quad \langle H_{\text{field}}\rangle_a=0.$$

It is interesting to compare this with the field due to two molecules in the uncorrelated state, $\psi_u=\varphi_1(1)\varphi_2(2)$, for which the initial energy is the same as for the two correlated states we have been considering. (All three states are eigenstates of the energy with the same eigenvalue.) According to Eq. (19), the field strength due to the molecules (both spontaneous and induced) is zero, and the part of the field energy due to the molecules [all but the first and last term of Eq. (18)] is

$$\langle H_{\text{field}}\rangle_u=\mathcal{E}_0(t),$$

just half the energy for the symmetric correlated state.

Since the expectation value of the field strength vanishes in all three cases, the molecular radiation, if any, must be incoherent spontaneous emission. We see, now, the meaning of quantum-mechanical correlation: In the correlated states, the incoherent oscillation of the molecules is correlated, being equal and in phase for the symmetric state, while equal and out of phase for the antisymmetric state. In other words, although

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[14] For purposes of the present article, we define any state which may be expressed in the form of Eq. (3) as uncorrelated. All other states are correlated in varying degree. A completely correlated energy state is an energy state which is itself a superpositon of linearly independent, uncorrelated energy states (obviously belonging to the same eigenvalue), with superposition coefficients satisfying certain relationships which need not be specified—for present purposes—in general form, since the aim is not to discuss the formalism of correlation (which is similar to the angular momentum formalism), but, rather, to point out certain salient features of simple correlated states which illustrate the effect of correlation. The correlated states discussed specifically in the following text are completely correlated. In justification of the foregoing definition, it may be pointed out that in a correlated state the result of an energy measurement on some molecule cannot be predicted with certainty, but this uncertainty is reduced as soon as an energy measurement is made on some (but not necessarily any) other molecule. In other words, the energy uncertainties for some (all, in the case of a completely correlated state) molecules are correlated. It goes without saying that this is a quantum-mechanical type of correlation and should not be confused with classical correlations (such as phase relationships between oscillating expectation values of the dipole moment, for instance) which may exist in an uncorrelated state.

[15] The two correlated states being considered are symmetric and antisymmetric, respectively. This has no relationship, however, to the Pauli principle, since the molecules are assumed to be sufficiently separated so that there is no overlap of spatial wave function.

we cannot predict the phase of oscillation of the molecules in a correlated energy state, the phase relationship between the molecules themselves is well defined. Here, again, it may be somewhat strange to label the same phenomenon by the words "incoherent" and "correlated," but there will be no difficulty if one bears in mind our definition of incoherence, namely, that the phase is an unpredictable (random) variable, as one examines similar "systems." In the case of uncorrelated states, the "system" is a single molecule, while in the case of correlated states, the "system" is the group of molecules which are correlated. (The word "system" is used here in a different sense than when setting up the Hamiltonian.)

We consider now a group of N molecules, and set up a correlated state with m molecules in the lower state and $N-m$ in the upper state. The particular correlation we choose is such that the wave function is symmetric with respect to the internal coordinates of all the molecules, and is the generalization of the symmetric state for two molecules considered above. We thus have

$$\psi_s = C\sum_P P\varphi_1(1)\cdots\varphi_1(m)\varphi_2(m+1)\cdots\varphi_2(N),$$

where P is the permutation operator and the summation is over all permutations which have the effect of interchanging (different) states of pairs of molecules. Since there are $N![m!(N-m)!]^{-1}$ such permutations,

$$C=\left(\frac{N!}{m!(N-m)!}\right)^{-\frac{1}{2}}.$$

We are interested in obtaining an expression for the spontaneous emission, so we assume $E_0=0$, for simplicity. The expectation value of the field strength, then, vanishes, since $\langle\gamma^{(0)}\rangle=0$. In evaluating the expectation value of the energy, we note that the only contribution of the molecules to $\langle P^2\rangle$ [see Eq. (17)] is through the term $\sum_{i,j}\langle P_i^{(1)}P_j^{(1)}\rangle$. The molecular operator in $P_m^{(1)}$ is $\gamma_m^{(0)}$, so that we need to calculate

$$\psi_s^*\left[\sum_{i,j}\gamma_i^{(0)}(t_1)\gamma_j^{(0)}(t_2)\right]\psi_s, \tag{28}$$

where t_1 and t_2 are variables of integration. Those operators in the summation for which $i=j$ connect only those terms in ψ_s and ψ_s^* which correspond to the same arrangement of states. There are thus

$$N\frac{N!}{m!(N-m)!} \tag{29}$$

such terms in (28). The operators $\gamma_i^{(0)}\gamma_j^{(0)}$, $i\neq j$, connect terms in ψ_s containing the factors $\varphi_2(i)\varphi_1(j)$ or $\varphi_1(i)\varphi_2(j)$ with the terms in ψ^* containing the factors $\varphi_1^*(i)\varphi_2^*(j)$ or $\varphi_2^*(i)\varphi_1^*(j)$, respectively. There are N^2-N such operators and each operator connects

$$2\frac{(N-2)!}{(m-1)!(N-m-1)!}$$

terms in ψ and ψ^*. There are, thus,

$$2(N^2-N)\frac{(N-2)!}{(m-1)!(N-m-1)!}=2m(N-m)C^{-2} \tag{30}$$

such terms in (31). Each one of these terms [referred to both in (29) and (30)] has the value $C^2\tilde{\gamma}^2\times\exp[\pm i\omega(t_1-t_2)]$, the sign of the exponent depending on i and j, and being of no consequence since t_1 and t_2 are variables of integration. Doing a similar calculation for the magnetic part of the field energy, we obtain, as the contribution of the molecules to the energy of the field,

$$\langle H_{\text{field}}\rangle_s=[\tfrac{1}{2}N+m(N-m)]\mathcal{E}_0(t).$$

This is a maximum when $m=\frac{1}{2}N$, for which we obtain an energy

$$(\tfrac{1}{4}N^2+\tfrac{1}{2}N)\mathcal{E}_0(t). \tag{31}$$

For an uncorrelated energy state with half the molecules in the upper state and half in the lower state, the spontaneous emission energy is, from Eq. (18),

$$\tfrac{1}{2}N\mathcal{E}_0(t).$$

Both of these energies are incoherent, and we see that for the correlated state chosen, the energy is proportional to N^2, while for the uncorrelated state it is proportional to N. This, of course, is due to the fact, as brought out in the discussion of the case of two molecules, that the molecular radiation, although not having, in this instance, a well defined phase, has the same phase for all the molecules in the case of the correlated state, and random phase relationships between molecules in the case of the uncorrelated state.

It is interesting to compare the above spontaneous emission energies with the spontaneous emission from N molecules which are oscillating coherently. Consider the state (3) with $|a_1(m)|=|a_2(m)|=2^{-\frac{1}{2}}$, $\theta_m=\theta$. This means that all the molecules are oscillating with the same (well-defined) phase and maximum amplitude. (Actually, in this state and the last two considered, the expectation value of the molecular energy is the same.) From Eq. (18) we have for this case a total spontaneous emission energy

$$\tfrac{1}{4}(N^2+N)\mathcal{E}_0(t). \tag{32}$$

Of this, as can be seen from Eqs. (20) and (21), the N^2 term is coherent, the electric field varying as $\sin(\omega t+\theta)$, and the N term is incoherent, having random phase. For large N, Eqs. (32) and (31) give the same energy, but it is important to note the physical difference between the type of radiation in the two cases. The incoherent radiation proportional to N^2 is due to a quantum-mechanical correlation between the molecules, all the molecules forming part of one "supermolecule," while the coherent radiation proportional to N^2 is the same as that coming from a collection of classical-type oscillators, all having the same phase.

It should be mentioned that there are other correlated states with the same number of molecules in the upper and lower states as the one considered above, but having different correlations. Some of these states correspond to the antisymmetric state we have considered for the case of two molecules; that is, the correlation is such that the radiation of the various molecules cancels out completely. And there are intermediate cases. Dicke[6] has considered these states in detail, and has called ψ_s, with $m\sim\frac{1}{2}N$, and those states which are almost like it, "super-radiant." He calls the radiation from "super-radiant" states "coherent" because the energy is proportional to N^2, but does not differentiate between radiation of well-defined phase and that of random phase.

One might ask if it is possible to superpose correlated states of different energies in order to obtain coherent oscillation. A simple illustration, using the case of two molecules again, will illuminate this problem. To the symmetric state of Eq. (26) we add amounts of the states $\varphi_1(1)\varphi_1(2)$ and $\varphi_2(1)\varphi_2(2)$ in the following manner:

$$\psi=A_1\varphi_1(1)\varphi_1(2)+A_2\varphi_2(1)\varphi_2(2)+A_s[\varphi_1(1)\varphi_2(2)+\varphi_2(1)\varphi_1(2)]. \quad (33)$$

Now, if we choose the coefficients in such a way as to obtain the maximum coherence, that is, maximum amplitude for the (coherently) oscillating dipole-moment expectation value, then it can be shown easily that Eq. (33) becomes a special case of Eq. (3), namely, an uncorrelated state. In other words, maximum coherence implies zero correlation. We have already shown that maximum correlation implies zero coherence, or incoherence. (Actually, we have shown this only for the symmetrically correlated state, ψ_s, but the same reasoning applies to any other completely correlated state.) Thus, coherence and correlation are mutually exclusive. Of course, it is a question of degree, and there can exist, in principle, some of both; but it can be shown, generally, for N molecules, that the more we have of correlation, the less we have of coherence, and vice versa. This also follows, generally, from the uncertainty principle.

The question now arises: in the physical situation in which the spontaneous emission is proportional to N^2, which is the correct description, one in terms of uncorrelated states or in terms of correlated states? A phenomenological answer to this question suggests itself immediately. If the phase of the spontaneous-emission field can be predicted, then it is the former, while if the phase is unpredictable, it is the latter. Another way of trying to answer the question is to examine the dynamics of the situation under consideration. One may assume initially an uncorrelated state (not an unreasonable assumption for a thermodynamic system; in the case of the ammonia beam maser, where there is state selection, we know exactly what to assume) and then let the dynamics tell us what happens. Actually, this is what we have done, except that we have not carried our analysis to high enough order to exhibit correlation effects. We note, for example, that $P^{(2)}$ [Eq. (15)] contains the operator $P^{(0)}\sum_m I_m$. The fourth-order term in the energy, therefore, will contain the operator $(P^{(0)})^2\sum_{m,m'}I_mI_{m'}$. This operator exhibits quantum-mechanical correlation effects, or correlation between the incoherent oscillations of the molecules, since $\langle(P^{(0)})^2\rangle$ does not vanish for $E_0=0$, and we obtain cross-terms for incoherent radiation. Similarly, it can be shown that there are other operators in the fourth- and higher-order terms of the energy which exhibit quantum-mechanical correlation effects. There are no such effects in terms of lower order than the fourth.

We apply the question posed in the last paragraph to the case of a large number of molecules excited by a strong rf pulse of short duration, a case which was treated earlier within the framework of uncorrelated states. Is the subsequent spontaneous emission coherent emission or incoherent correlated emission? It should be noted that in both instances the energy emitted and the instantaneous emission rate may be the same, as can be seen from Eqs. (31) and (32). From the above discussion it is clear that quantum-mechanical correlation will come about (if it does not exist initially) from the interaction of the molecules with one another through their radiation. An externally imposed field will not bring about such correlation directly. On the other hand, it *will* generate an (oscillating) electric dipole moment in each molecule, the expectation value of which bears a definite phase relationship to the field. We may conclude, therefore, that the subsequent spontaneous emission is of the coherent type possible with uncorrelated states. The well-known fact that the spontaneous emission bears a definite phase relationship to the exciting field indicates that our conclusion is correct, and we were therefore justified in treating the problem by means of uncorrelated states, in part III.

For the sake of simplicity we have assumed all along that the molecules are all exposed to the field at time $t=0$, which is also (for the initial state of the field which we have assumed) the time at which the phase of the expectation value of the field is zero. This assumption of special entrance phase is no limitation, however, since we have considered times such that $t\gg\omega^{-1}$, and have considered only secular effects (such as changes in the amplitude of oscillation occurring during times long compared to a cycle) which are independent of the entrance phase, discarding terms which are essentially transient effects, depending on the entrance phase, and significant only during the first few cycles.

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