Coulomb potential[17] or that based on the TF approximation.[16] It is expected that the results of these calculations will be reported in detail in a later paper.

On the basis of these results, it may then be of interest to calculate atomic scattering cross sections (in suitable energy intervals), and to perform range-energy calculations, both capable of being verified by experi-

ment.[18-20] Furthermore, the TF three-center potential (115) may be applied to suitable triatoms. Lastly, a generalization of the formalism developed in this paper to *ionic* systems would evidently still further enhance its usefulness.

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Onset of Correlation in Initially Uncorrelated System

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The system under consideration is a number of molecules contained in a resonant cavity and isolated from external influences. The molecules are assumed to have two energy levels, and the molecular frequencies have a Gaussian distribution centered at the cavity frequency; the initial states of the molecules are uncorrelated. The onset of correlation in the molecular behavior is studied by examining the field in the cavity and the power emitted by the molecules for effects depending on the square of the number of molecules, in a perturbation theory approach.

It is shown that correlation effects manifest themselves in the fourth order interaction. Both the correlation energy in the field and the correlation power emitted by the molecules approach steady-state values after transient periods determined by the relaxation time of the cavity and the frequency spread of the molecules. A physical picture of the correlation effects, as being due to induced emission produced by the lowest order spontaneous and thermal emission, is investigated and found to be approximately correct. The ratio of correlation energy to lowest order spontaneous emission energy is derived. An analysis is made of the dependence of the results on the initial states of the molecules, and interpreted in terms of the physical picture. The effect of the presence of a number of cavity modes, rather than a single mode, within the frequency spread of the molecules is investigated under simplifying assumption, and is shown to multiply the correlation effects by the square of the number of modes.

INTRODUCTION

THE subject of correlation in a many-body system includes a large number of diverse problems, some of which have become very popular recently. Correlation is caused, of course, by a coupling between the individual bodies of the system, which can take one one or more of several forms. The problem to be treated in the present article deals with the correlation of a number of atomic systems, which we refer to as molecules, coupled to one another through the electromagnetic field in a resonant cavity. The question we ask is the following: Suppose we have a cavity containing a number of molecules that are isolated from external influences and are initially in uncorrelated states. Will correlation arise, and if so, in what manner?

The meaning of correlation in the present context will be described first. The molecules are uncorrelated if the state of each molecule is independent of the other molecules. Correlation may be measured by the extent to which the behavior of each molecule is affected by the others. The difference between the behavior of molecules in correlated and uncorrelated states has been discussed in some detail by both Dicke[1] and the author.[2] In reference 2 it is shown that when molecules

are in a correlated state, spontaneous emission is proportional to N^2, where N is the number of molecules; if the molecules are in an uncorrelated state that is also an energy state, spontaneous emission is proportional to N. There are, however, uncorrelated states in which each molecule is in the same superposition of individual energy states, and in this case the spontaneous emission is also proportional to N^2. The latter type of state may be regarded as corresponding to a classical array of dipoles oscillating with the same well-defined phase; it can be created by subjecting the molecules to an external driving field. We are excluding the consideration of such classical-type correlations, since they cannot arise in an isolated system, and use the term correlation to indicate only quantum-mechanical correlation. (In the language of reference 2, the case in which each molecule is in the same superposition of individual energy states is called uncorrelated but coherent.) Incidentally, as explained in reference 2, in the correlated energy state the molecules may also be regarded as oscillating in phase with each other, but the absolute phase is completely undetermined.[3] With

[1] R. H. Dicke, Phys. Rev. 93, 99 (1954).
[2] I. R. Senitzky, Phys. Rev. 111, 3 (1958).

[3] We give a simple illustration of correlated and uncorrelated states for the case of two similar molecules. If φ_{mi} is the i'th energy state of the mth molecule, then $\varphi_{1i}\varphi_{2j}$ is an uncorrelated energy state, and $2^{-\frac{1}{2}}(\varphi_{1i}\varphi_{2j} \pm \varphi_{1j}\varphi_{2i})$ is a correlated energy state. In the language of reference 2, $\psi_1\psi_2$, where $\psi_m = a_1\varphi_{m1} + a_2\varphi_{m2}$, is a coherent uncorrelated state.

the above exclusion, we can regard emission proportional to N^2 as an indication of correlation.

The problem of the present article may now be formulated precisely. We consider a number of initially uncorrelated molecules in a cavity. There are no external influences (such as driving field) acting on the system. We study both the field in the cavity and the energy of the molecules as a function of time; the quantities in which we are particularly interested are those proportional to N^2.

The equations of motion and other preliminaries are discussed in Part I. Expressions for the field energy are obtained in Part II, and the physical significance of these expressions is analyzed in Part III. In Part IV the power radiated by the molecules is examined. In Part V the discussion is extended to the case in which a number of cavity modes, rather than a single mode, interact with the molecules.

I

The molecules and the cavity are similar in their properties to those considered in a recent investigation of induced and spontaneous emission in a coherent field.[4] Each molecule has two energy states and a molecular (angular) frequency

$$\omega_m = (E_{m2} - E_{m1})/\hbar. \tag{1}$$

The spread in molecular frequencies is Gaussian, so that the number of molecules per unit frequency range at ω_m is given by

$$\rho(\omega_m) = N\alpha\pi^{-\frac{1}{2}}\exp[-\alpha^2(\omega_m - \omega)^2], \tag{2}$$

ω being the frequency of the single cavity mode under consideration. The loss in the cavity walls is described by the loss constant β, which is specified in terms of the cavity Q

$$\beta = \omega/Q_c. \tag{3}$$

The molecules are coupled to the cavity field by an electric dipole moment $\boldsymbol{\gamma}$ having only off-diagonal matrix elements (no static dipole moment). The electromagnetic field in the cavity is described in the usual manner by

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

where $\mathbf{u}(\mathbf{r})$ is a normalized function describing the spatial dependence of the field in the cavity, and $Q(t)$ and $P(t)$ are the quantum-mechanical field operators satisfying the commutation relationship $[Q(t),P(t)]$

$= i\hbar$. The method of introducing loss into a quantum-mechanical formalism was discussed in detail in two previous articles,[5,6] and only the results will be restated here, as necessary.

We begin with the Hamiltonian

$$H = H_f + H_l + H_{fl} + \sum_m H_m + \sum_m 4\pi cu_m\gamma_m P, \tag{5}$$

where H_f is the Hamiltonian of the electromagnetic field, H_l is the Hamiltonian of the loss mechanism, H_{fl} is the term describing the coupling between field and loss mechanism, and H_m is the Hamiltonian of the mth molecule. The last term describes the coupling between the molecules and the field, u_m representing the magnitude of $\mathbf{u}(\mathbf{r}_m)$, and γ_m the component of $\boldsymbol{\gamma}$ along $\mathbf{u}(\mathbf{r}_m)$. The only individual Hamiltonian which need be given explicitly in terms of coordinates and momenta is

$$H_f = 2\pi c^2 P^2 + (\omega^2/8\pi c^2)Q^2. \tag{6}$$

In reference 6 it is shown that the equation of motion for P is

$$\ddot{P} + \beta\dot{P} + \omega^2 P = F(t) - (\omega^2/c)\sum_m u_m\gamma_m(t), \tag{7}$$

where $F(t)$ is an operator which expresses both the thermal and quantum-mechanical fluctuation properties of the loss mechanism:

$$\langle F(t)\rangle = 0, \tag{8}$$

$$\langle F(t_1)F(t_2)\rangle = \frac{\omega^3\hbar\beta}{4\pi^2 c^2}\left[i\frac{\mathcal{P}}{t_1 - t_2} + \pi\delta(t_1 - t_2)f(T)\right], \tag{9}$$

$$f(T) \equiv [1 + 2(e^{\hbar\omega/kT} - 1)^{-1}], \tag{10}$$

T being the temperature of the cavity. We do not need a similar equation of motion for Q, since if P is known Q can be obtained from

$$Q = -(4\pi c^2/\omega^2)\dot{P}, \tag{11}$$

which follows from Eqs. (5) and (6). The remaining equations of motion follow immediately from the Hamiltonian of Eq. (5):

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

$$i\hbar\dot{H}_m(t) = 4\pi cu_m P(t)[H_m(t), \gamma_m(t)]. \tag{13}$$

It will be more convenient for our purposes to recast the differential equations of motion into integral equations. One can verify by differentiation that

$$H_m(t) = H_m{}^{[0]} + \frac{2\pi cu_m}{i\hbar}\int_0^t dt_1\{[H_m(t_1),\gamma_m(t_1)],P(t_1)\}, \tag{14}$$

$$\gamma_m(t) = \gamma_m{}^{[0]}(t) + \frac{4\pi cu_m}{\hbar^2}\int_0^t dt_1\int_0^{t_1} dt_2\, U_m(t-t_1)[\gamma_m(t_1),[\gamma_m(t_2),H_m(t_2)]P(t_2)]U_m{}^{-1}(t-t_1), \tag{15}$$

[4] I. R. Senitzky, Phys. Rev. 119, 1807 (1960). The term "spontaneous emission" should be understood as referring to the lowest order effect of the molecules. Higher order effects, which are the subject of the present article, are referred to as "correlation" effects, for clarity.

[5] I. R. Senitzky, Phys. Rev. 115, 227 (1959).

[6] I. R. Senitzky, Phys. Rev. 119, 670 (1960). Note the change of notation from $D(t)$ in this reference to $F(t)$ in the present article.

where

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

$$\gamma_m^{[0]}(t) = \tilde{\gamma}\begin{pmatrix} 0 & e^{-i\omega_m t} \\ e^{i\omega_m t} & 0 \end{pmatrix}, \quad H_m^{[0]} = \begin{pmatrix} E_{m1} & 0 \\ 0 & E_{m2} \end{pmatrix},$$

and where the symmetrized product $(\{A,B\} \equiv AB + BA)$ is used for convenience. The operators $H_m^{[0]}$ and $\gamma_m^{[0]}(t)$ describe the dipole moment[7] and energy of the mth molecule in the absence of coupling to the field, and are the zeroth order quantities in a perturbation calculation. These expressions also yield the initial values in the presence of the coupling. We see that the coupling between molecules and field may be considered to be turned on at $t=0$. As far as the integral equation for the field is concerned, it is more realistic to consider the coupling between the field and loss mechanism to be turned on at $t = -\infty$. We then have, from Eq. (7) and the condition $\beta/\omega \ll 1$,

$$P = P_F + P_\gamma, \tag{16}$$

where

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

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

P_F is obviously the zeroth order solution for $P(t)$. Equations (14), (15), and (16) are three integral equations for the three operators P, γ_m, H_m. They are of a form which is suitable for an iterative type of calculation in perturbation theory, the zeroth-order solutions being given explicitly.

The quantities to which we will attach physical meaning are expectation values. We will calculate the expectation values of the field energy in the cavity and the molecular energy. In order to perform this calculation in the Heisenberg picture, we must specify the initial state of the molecules. As in reference 4, we may want to consider several possible initial states, such as a thermal state or a state in which all the molecules have the same energy. The state of each molecule is given, in general, by

$$\psi_m = a_{m1}\varphi_{m1} + a_{m2}\varphi_{m2},$$

and the state of the group of molecules is described by

$$\Psi = \prod_{m=1}^{N} \psi_m.$$

In all the situations to be considered, we assume that the initial phase difference between the two super-

position constants a_{m1} and a_{m2}, which we call the phase of the molecule, is a random function of m, the molecular label. This randomness implies that we are dealing with a number of molecules that have not been prepared beforehand to oscillate with the same phase. We will therefore drop those terms in the expectation value which contain a summation of the initial phases of the molecules. This means that only diagonal terms of the operators under consideration, in the energy representation of the (free) molecules, will contribute to the expectation value.

Our problem can now be narrowed. We are looking for the lowest-order terms in the field energy and the molecular energy operators which (1) are proportional to N^2; (2) contain diagonal elements with respect to the molecules, and (3) are not linear in F. The first condition is our criterion for correlation, the second condition has just been explained, and the third condition follows from Eq. (8).

II

The expectation value of the field energy is given by

$$\langle H_f \rangle = 2\pi c^2 \langle P^2 \rangle + (\omega^2/8\pi c^2)\langle Q^2 \rangle. \tag{17}$$

The evaluations which we must make here, as well as in the later sections, are fairly complicated and lengthy. In order to illustrate the method used, we will go into detail only in the evaluation of $\langle P^2 \rangle$.

If we express the higher-order operators in terms of the unperturbed operators, P_F, $\gamma_m^{[0]}$, and $H_m^{[0]}$, we obtain, in general, a sum of numerous terms. Only a few of the many terms of P^2 will satisfy our three conditions. It is therefore advisable to develop a scheme by which it is possible to pick out for computation only those terms contributing toward our final result. We note that in order to obtain proportionality to N^2, we need at least a double summation. Whether or not a term contains diagonal elements with respect to the mth molecule depends on the number of times the factor $\gamma_m^{[0]}$ occurs in that term, since $H_m^{[0]}$ is a diagonal matrix and does not alter the diagonality properties of the term. If $\gamma_m^{[0]}$ occurs an even number of times, the matrix is diagonal; and if it occurs an odd number of times, the matrix is off-diagonal. The arguments of the operators, the commutator and symmetrizing brackets, the integrals, and the numerical coefficients are not pertinent so far as the three conditions are concerned. We can therefore use, for each term, a shorthand notation that omits arguments, brackets, integrals, and numerical coefficients. $H_m^{[0]}$ may also be dropped. It is important, however, to retain the summation signs.

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[7] Strictly speaking, $\tilde{\gamma}$ should be labeled $\tilde{\gamma}_m$, since it gives the amplitude of the component of dipole moment along $u(r_m)$. However, since there is no correlation between the polarization of the molecules and the direction of the field, we can consider an average $\tilde{\gamma}_m$ denoted by $\tilde{\gamma}$.

We use an arrow instead of an equal sign to indicate correspondence of the shorthand symbol. For further simplicity we drop the superscripts (which indicate the order) of the zeroth order quantities in the shorthand term. We thus have

$$P^{[0]} \rightarrow F, \tag{18}$$

$$P^{[1]} \rightarrow \sum \gamma_m, \tag{19}$$

$$\gamma_m{}^{[1]} \rightarrow \gamma_m{}^2 F, \tag{20}$$

$$H_m{}^{[1]} \rightarrow \gamma_m F, \tag{21}$$

$$P^{[2]} \rightarrow \sum \gamma_m{}^2 F, \tag{22}$$

$$\gamma_m{}^{[2]} \rightarrow \gamma_m \gamma_m{}^{[1]} F + \gamma_m{}^2 P^{[1]} + \gamma_m{}^3 F^2,$$
$$\rightarrow \sum_{m'} \gamma_m{}^2 \gamma_{m'} + \gamma_m{}^3 F^2, \tag{23}$$

$$P^{[3]} \rightarrow \sum_{m,m'} \gamma_m{}^2 \gamma_{m'} + \sum_m \gamma_m{}^3 F^2. \tag{24}$$

It can be seen very easily from these expressions that no order lower than the fourth in an expression for P^2 will fulfill the three conditions enumerated above. We therefore select the terms in $P^{2[4]}$ that satisfy the conditions.

One sees immediately that $P^{[2]2}$, for which the shorthand symbol is $\sum_{m,m'} \gamma_m{}^2 \gamma_{m'}{}^2 F^2$, meets the requirements. So does the term in $\{P^{[1]}, P^{[3]}\}$, which is described by the short-hand symbol $\sum_{m,m',m''} \gamma_m \gamma_{m'}{}^2 \times \gamma_{m''}$, when we take only that part of the summation for which $m'' = m$. In a similar manner we can select the pertinent term in $P^{[0]} P^{[4]}$ after we have obtained the shorthand symbol for $P^{[4]}$, which is rather lengthy. We write now those terms contributing toward that part of $P^{2[4]}$ which meets our requirements. Where the expression for an operator has several terms, and where some terms are dropped because they do not contribute toward the result in which we are interested, we put the symbol $\wedge$ over the operator to indicate that the expression is incomplete, but contains all that we can use. We have

$$P^{[1]}(t) = -\frac{\omega}{c} \sum_m u_m \int_0^t dt_1 \, \gamma_m{}^{[0]}(t_1) \varphi(t - t_1), \tag{25}$$

where

$$\varphi(\tau) \equiv e^{-\frac{1}{2}\beta\tau} \sin\omega\tau;$$

$$P^{[2]}(t) = -\frac{8\pi}{\hbar} \omega\bar{\gamma}^2 \sum_m u_m{}^2 I_m \int_0^t dt_1 \int_0^{t_1} dt_2 \, \varphi(t - t_1) P_F(t_2) \sin\omega_m(t_1 - t_2), \tag{26}$$

where

$$I_m \equiv \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}_m,$$

the subscript m indicating that I_m operates on the state vector of mth molecule only;

$$\hat{P}^{[3]}(t) = \frac{4\pi\omega^2}{c\hbar^2} \sum_{m,m'} u_m{}^2 u_{m'} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4$$

$$\times \varphi(t - t_1)\varphi(t_3 - t_4) U_m(t_1 - t_2)[\gamma_m{}^{[0]}(t_2), [\gamma_m{}^{[0]}(t_3), H_m{}^{[0]}]\gamma_{m'}{}^{[0]}(t_4)] U_m{}^{-1}(t_1 - t_2); \tag{27}$$

$$\hat{P}^{[4]}(t) = -\frac{4\pi\omega}{c\hbar^2} \sum_m u_m{}^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \, \varphi(t - t_1)[\gamma_m{}^{[0]}(t_2), [\gamma_m{}^{[0]}(t_3), H_m{}^{[0]}]P^{[2]}(t_3)],$$

$$= \frac{32\pi^2}{c\hbar^3} \omega^2\bar{\gamma}^2 \sum_{m,m'} u_m{}^2 u_{m'}{}^2 \int_0^t dt_1 \cdots \int_0^{t_4} dt_5 \, \varphi(t - t_1)\varphi(t_3 - t_4) \sin\omega_{m'}(t_4 - t_5) P_F(t_5)$$

$$\times [\gamma_m{}^{[0]}(t_2), [\gamma_m{}^{[0]}(t_3), H_m{}^{[0]}]I_{m'}]. \tag{28}$$

Since we are interested only in effects depending on N^2 and not on N, we ignore the $m = m'$ terms in Eqs. (27) and (28). This allows us to take the m' factor out of the commutator brackets and obtain simpler expressions:

$$\hat{P}^{[3]}(t) \approx \frac{8\pi}{c\hbar} \omega^2\bar{\gamma}^2 \sum_{m \neq m'} u_m{}^2 u_{m'} I_m \omega_m \int_0^t dt_1 \cdots \int_0^{t_3} dt_4 \, \varphi(t - t_1)\varphi(t_3 - t_4)\gamma_{m'}{}^{[0]}(t_4) \cos\omega_m(t_2 - t_3)$$

$$= \frac{8\pi}{c\hbar} \omega^2\bar{\gamma}^2 \sum_{m \neq m'} u_m{}^2 u_{m'} I_m \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \, \varphi(t - t_1)\varphi(t_2 - t_3)\varphi_{m'}{}^{[0]}(t_3) \sin\omega_m(t_1 - t_2), \tag{29}$$

$$\hat{P}^{[4]}(t) \approx \frac{64\pi^2}{\hbar^2}\omega^2\tilde{\gamma}^4 \sum_{m\neq m'} u_m{}^2 u_{m'}{}^2 \omega_m I_m I_{m'} \int_0^t dt_1 \cdots \int_0^{t_4} dt_5\, P_F(t_5)\, \varphi(t-t_1)\, \varphi(t_3-t_4)\, \sin\omega_{m'}(t_4-t_5)\, \cos\omega_m(t_2-t_3)$$

$$= \frac{64\pi^2}{\hbar^2}\omega^2\tilde{\gamma}^4 \sum_{m\neq m'} u_m{}^2 u_{m'}{}^2 I_m I_{m'} \int_0^t dt_1 \cdots \int_0^{t_3} dt_4\, P^{[0]}(t_4)\, \varphi(t-t_1)\, \varphi(t_2-t_3)\, \sin\omega_m(t_1-t_2)\, \sin\omega_{m'}(t_3-t_4). \qquad (30)$$

We consider now the products occurring in $\langle P^{2[4]}\rangle$. From Eq. (26)

$$\langle P^{[2]2}\rangle = \frac{32\pi^2}{\hbar^2}\omega^2\tilde{\gamma}^4 \sum_{m,m'} u_m{}^2 u_{m'}{}^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^t dt_3 \int_0^{t_3} dt_4\, \sin\omega_m(t_1-t_2)\, \sin\omega_{m'}(t_3-t_4)\, \varphi(t-t_1)\, \varphi(t-t_3)$$
$$\times \langle\{I_m P_F(t_2),\, I_{m'} P_F(t_4)\}\rangle. \qquad (31)$$

Again, ignoring the $m=m'$ terms, we have

$$\langle P^{[2]2}\rangle = \frac{32\pi^2}{\hbar^2}\omega^2\tilde{\gamma}^4 \sum_{m\neq m'} u_m{}^2 u_{m'}{}^2 \langle I_m\rangle\langle I_{m'}\rangle \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^t dt_3 \int_0^{t_3} dt_4\, \sin\omega_m(t_1-t_2)\, \sin\omega_{m'}(t_3-t_4)$$
$$\times \varphi(t-t_1)\, \varphi(t-t_3)\, \langle\{P_F(t_2),\, P_F(t_4)\}\rangle. \qquad (32)$$

We convert the summations over m and m' to integrations over ω_m and $\omega_{m'}$. Assuming that $u_m{}^2$ and $\langle I_m\rangle$ are sufficiently slowly varying functions of ω_m to be taken outside of the integral sign as averages, which we call u^2 and $\langle I\rangle$, respectively, and making use of Eq. (2), we obtain

$$\langle P^{[2]2}\rangle = \frac{32\pi^2}{\hbar^2}\omega^2\tilde{\gamma}^4 N^2 u^4 \langle I\rangle^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^t dt_3 \int_0^{t_3} dt_4 \langle\{P_F(t_2),\, P_F(t_4)\}\rangle\, \varphi(t-t_1)\, \varphi(t-t_3)\, \sin\omega(t_1-t_2)$$
$$\times \sin\omega(t_3-t_4)\, \exp\{-(1/4\alpha^2)[(t_1-t_2)^2+(t_3-t_4)^2]\}. \qquad (33)$$

The evaluation of $\langle\{P_F(t_2),P_F(t_4)\}\rangle$ is carried out by means of Eq. (9). It is shown in Appendix A that

$$\langle\{P_F(t_2),P_F(t_4)\}\rangle = (\hbar\omega/4\pi c^2) f(T) e^{-\frac{1}{2}\beta|t_2-t_4|}\, \cos\omega(t_2-t_4). \qquad (34)$$

We have, thus

$$\langle P^{[2]2}\rangle = \frac{8\pi}{c^2\hbar}\omega^3\tilde{\gamma}^4 u^4 N^2 \langle I\rangle^2 f(T) \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^t dt_3 \int_0^{t_3} dt_4 E\, \exp[-\tfrac{1}{2}\beta(|t_2-t_4|+2t-t_1-t_3)]\, \sin\omega(t-t_1)$$
$$\times \sin\omega(t-t_3)\, \sin\omega(t_1-t_2)\, \sin\omega(t_3-t_4)\, \cos\omega(t_2-t_4), \qquad (35)$$

where

$$E \equiv \exp\{-(1/4\alpha^2)[(t_1-t_2)^2+(t_3-t_4)^2]\}.$$

The product of trigonometric functions in the integrand of Eq. (35) is equal to a constant plus oscillatory terms. We drop the oscillatory terms and retain only the constant, which is $\frac{1}{16}$. Our expression for $\langle P^{[2]2}\rangle$ is now

$$\langle P^{[2]2}\rangle = \tfrac{1}{2}A \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^t dt_3 \int_0^{t_3} dt_4 E\, \exp[-\tfrac{1}{2}\beta(2t-t_1-t_3+|t_2-t_4|)], \qquad (36)$$

where

$$A \equiv (\pi/c^2\hbar)\omega^3\tilde{\gamma}^4 u^4 N^2 \langle I\rangle^2 f(T).$$

We deal with the absolute value in the exponent by dividing the region of integration into two regions: one in which $t_2 \leqslant t_4$, and the other in which $t_2 > t_4$. Thus

$$\int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^t dt_3 \int_0^{t_3} dt_4 E\, \exp[-\tfrac{1}{2}\beta(2t-t_1-t_3+|t_2-t_4|)]$$
$$= \int_0^t dt_2 \int_0^t dt_4 \int_{t_2}^t dt_1 \int_{t_4}^t dt_3 E\, \exp[-\tfrac{1}{2}\beta(2t-t_1-t_3+|t_2-t_4|)]$$
$$= Y+Z, \qquad (37)$$

where

$$Y \equiv \int_0^t dt_2 \int_0^{t_2} dt_4 \int_{t_2}^t dt_1 \int_{t_4}^t dt_3 E\, \exp[-\tfrac{1}{2}\beta(2t-t_1+t_2-t_3-t_4)],$$

and

$$Z = \int_0^t dt_2 \int_t^t dt_4 \int_{t_2}^t dt_1 \int_{t_4}^t dt_3 \, E \exp[-\tfrac{1}{2}\beta(2t - t_1 - t_2 - t_3 + t_4)].$$

By interchanging t_2 with t_4 and t_3 with t_1, we see easily that

$$Z = \int_0^t dt_4 \int_{t_4}^t dt_2 \int_{t_4}^t dt_3 \int_{t_2}^t dt_1 \, EJ$$

$$= \int_0^t dt_2 \int_0^{t_2} dt_4 \int_{t_2}^t dt_1 \int_{t_4}^t dt_3 \, EJ$$

$$= Y, \tag{38}$$

where

$$J \equiv \exp[-\tfrac{1}{2}\beta(2t - t_1 + t_2 - t_3 - t_4)].$$

Thus

$$\langle P^{[2]2} \rangle = A Y. \tag{39}$$

For later application, it is convenient to recast Y. By rearranging the order of integration, and interchanging t_3 with t_4 (which leaves the integrand of Y invariant), we have

$$Y = \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^t dt_4 \, EJ - \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \, EJ. \tag{40}$$

Using methods similar to those with which we derived Eq. (39) from Eq. (31), we obtain

$$\langle \{P^{[0]}(t), P^{[4]}(t)\} \rangle = A \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \, EJ, \tag{41}$$

and

$$\langle \{P^{[1]}, P^{[3]}\} \rangle = B \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^t dt_4 \, EJ, \tag{42}$$

where

$$B \equiv (\pi/c^2\hbar)\omega^3\bar{\gamma}^4 u^4 N^2 \langle I \rangle.$$

From Eqs. (39), (40), (41), and (42) we finally have

$$\langle P^{2[4]} \rangle = B[1 + f(T)\langle I \rangle] \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^t dt_4 \, EJ. \tag{43}$$

We can calculate $\langle Q^{2[4]} \rangle$ in the same manner as $\langle P^{2[4]} \rangle$, or we can make use of the fact that (when averaged over a cycle) the electric energy in the cavity is approximately equal to the magnetic energy, provided $(\beta/\omega) \ll 1$ and the molecular coupling energy is small. We therefore have, from Eq. (6)

$$\langle H_f^{[4]} \rangle = (4\pi^2/\hbar)N^2\omega^3\bar{\gamma}^4 u^4 \langle I \rangle[1 + f(T)\langle I \rangle] X(t), \tag{44}$$

where

$$X(t) \equiv \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^t dt_4 \, EJ.$$

There remains now the task of evaluating the integral in Eq. (44). It obviously cannot be evaluated in closed form, since it contains Gaussian exponentials in the integrand. However, it can be partly integrated, as shown in Appendix B, to yield

$$X(t) = \frac{1}{\beta^2}\left\{ \int_0^t d\eta \int_0^{t-\eta} d\eta' \, E(\eta,\eta')e^{-\frac{1}{2}\beta(\eta+\eta')} + \int_0^t d\eta \int_0^\eta d\eta' \, E(\eta,\eta')[2 + \beta(\eta - \eta')]e^{-\frac{1}{2}\beta(\eta-\eta')} \right\}$$

$$+ \frac{1}{\beta^2}e^{-\beta t}\left\{ \int_0^t d\eta \int_0^{t-\eta} d\eta' \, E(\eta,\eta')[\beta(\eta+\eta'-t)-1]e^{\frac{1}{2}\beta(\eta+\eta')} + \int_0^t d\eta \int_0^t d\eta' \, E(\eta,\eta')[\beta(\eta-t)-1]e^{\frac{1}{2}\beta(\eta+\eta')} \right\}, \tag{45}$$

where

$$E(\eta,\eta') = \exp[-(1/4\alpha^2)(\eta^2+\eta'^2)].$$

III

We discuss now some interesting aspects of the correlation energy given by Eqs. (44) and (45). Perhaps the most interesting aspect is its approach to a steady-state value[8] after a transient period, which is determined by two time constants: the cavity relaxation time β^{-1} and the inverse molecular frequency width $\frac{1}{2}\alpha$. The steady-state value of X may be written, after some transformation, as

$$X_{st} = \frac{4}{\delta_c^2 \delta_m^2}\left\{\pi \exp(2r^2)(1-\mathrm{erf}r)^2 + \int_0^\infty dy \int_y^\infty dx\right.$$
$$\left.\times \exp[-\tfrac{1}{8}(x^2+y^2)](1+ry)e^{-ry}\right\}, \quad (46)$$

where

$$\mathrm{erf}r \equiv 2\pi^{-\frac{1}{2}}\int_0^r \exp(-x^2)dx,$$

and where, instead of using α and β, we have introduced the molecular and cavity frequency widths

$$\delta_c \equiv \beta, \quad \delta_m \equiv 2/\alpha, \quad (47)$$

and their ratio

$$r \equiv \delta_c/\delta_m.$$

We also define for convenience the molecular Q,

$$Q_m \equiv \omega/\delta_m. \quad (48)$$

The expression for X_{st} may be evaluated explicitly in the limits of small and large r, respectively. It is shown in Appendix C that in the limit of small r

$$X_{st} \to 8\pi/\delta_c^2 \delta_m^2, \quad (49)$$

so that

$$\langle H_f^{[4]}\rangle_{st} \approx (32\pi^3/\hbar\omega)N^2 Q_c^2 Q_m^2 \bar\gamma^4 u^4 \langle I\rangle$$
$$\times[1+f(T)\langle I\rangle], \quad (50)$$

and in the limit of large r

$$X_{st} \to 8(2\pi)^{\frac{1}{2}}/\delta_c^3 \delta_m, \quad (51)$$

so that

$$\langle H_f^{[4]}\rangle_{st} \approx [32(2\pi^5)^{\frac{1}{2}}/\hbar\omega]N^2 Q_c^3 Q_m \bar\gamma^4 u^4 \langle I\rangle$$
$$\times[1+f(T)\langle I\rangle]. \quad (52)$$

We see that as the molecular frequency spread becomes infinitesimal, the steady-state correlation energy becomes infinite. This merely means that there is no steady state for the case in which all the molecules are in resonance with the cavity. In fact, an examination of the first curly bracket in Eq. (45) shows that the second integral (the same one which, in the steady state,

becomes infinite with r) gives rise to a term linear in t when $\delta_m=0$ (or $\alpha=\infty$).

We can obtain an insight into the physical meaning of the correlation energy by considering at first, the factor

$$N^2\langle I\rangle[1+\langle I\rangle], \quad (53)$$

which is obtained from Eq. (44) by setting $T=0$. It is shown in reference 4 that the spontaneous-emission energy due to a single molecule is proportional to $[1+\langle I\rangle]$ and the induced-emission energy is proportional to $\langle I\rangle$ times the energy of the driving field. If we consider therefore, the induced emission of one molecule produced by the spontaneous emission of all the other molecules, and multiply by the total number of molecules, we obtain just the factor (53), remembering, of course, that we are neglecting terms in N compared to terms in N^2. We have, therefore, a tentative interpretation of the origin of the correlation energy.

It is interesting to carry this physical interpretation further and to make a calculation based on it. One may expect that this interpretation will have greater applicability during the steady-state condition than during the transient period. The results of reference 4 for the steady-state condition may be summarized as follows: Labeling the expectation value of the (zero order) thermal energy in the field [which is $\hbar\omega(e^{\hbar\omega/kT}-1)^{-1}$] by E_T, and labeling the coherent driving field energy in the cavity by E_D, we have the thermally-induced energy given by

$$E_T\epsilon\langle I\rangle R, \quad (54)$$

the spontaneously-emitted energy of the molecules given by

$$\tfrac{1}{2}\hbar\omega\epsilon(1+\langle I\rangle)R, \quad (55)$$

and the induced energy due to the coherent driving field given by

$$E_D\epsilon\langle I\rangle, \quad (56)$$

where

$$\epsilon \equiv (16\pi^{\frac{3}{2}}/\hbar\omega)NQ_c Q_m u^2 \bar\gamma^2,$$

and

$$R \equiv \tfrac{1}{2}\exp(r^2)(1-\mathrm{erf}r).$$

We note that when the driving field is coherent, one obtains the induced energy from the driving energy by multiplying by the factor $\epsilon\langle I\rangle$; and when the driving field is incoherent in the manner of the thermal field, we obtain the induced energy from the driving energy by multiplying by the factor $\epsilon\langle I\rangle R$. The spontaneous-emission energy is similar in its coherence properties to the thermal energy. Therefore, one might expect, at first glance, that the factor $\epsilon\langle I\rangle R$ should be used in order to obtain the correlation energy from the spontaneous-emission energy. Further thought, however, leads to the realization that this reasoning amounts to the breaking up of a higher order process into a succession of *independent* lower order processes, which is, in general, incorrect. Thus, the probability of a

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[8] It should be remembered, of course, that the time for which the present theory is valid is restricted by the use of perturbation theory; that is, it must be short compared to the time required for a substantial change of the state of the system.

higher order transition is generally not equal to the product of the probabilities of the constituent lower-order transitions, since correlation (commonly called coherence) effects are neglected. We try to incorporate these effects, in an intuitive manner, by using the factor which gives the induced emission for a *coherent* field. Furthermore, since the spontaneous-emission energy and thermally-induced energy have similar properties, we combine them and consider the effect of their sum,[9]

$$[\tfrac{1}{2}\hbar\omega(1+\langle I\rangle)+E_T\langle I\rangle]\epsilon R=\tfrac{1}{2}\hbar\omega[1+f(T)\langle I\rangle]\epsilon R, \quad (57)$$

in inducing emission. To derive our result, we merely multiply this energy by $\epsilon\langle I\rangle$, obtaining

$$(128\pi^3/\hbar\omega)N^2Q_c{}^2Q_m{}^2\bar{\gamma}^4u^4\langle I\rangle[1+\langle I\rangle f(T)]R. \quad (58)$$

This expression must now be compared with Eqs. (44) and (46), which give the steady-state value of the correlation energy. Since we do not have an evaluation of the second integral in Eq. (46), we look at the limiting values of the correlation energy for r large and small, given by Eqs. (52) and (50), respectively. Using the asymptotic value of the error function found in Appendix C, we note that

$$R\rightarrow\tfrac{1}{2}r^{-1}\pi^{-\frac{1}{2}}. \quad (59)$$

Substituting in Eq. (58) and comparing with Eq. (52), we see that for r large our intuitively derived expression is just $\sqrt{2}$ times the correct expression. As r becomes small, the error function approaches zero. Comparing Eq. (58) in the limit of small r with Eq. (50), we see that in this limit our intuitively derived expression is two times the correct expression. These results indicate that our intuitive interpretation of the correlation energy, as the induced-emission energy produced by the (lowest order) spontaneous and thermal-emission energy, is reasonable.[10]

A quantity which is of interest in connection with the present discussion, particularly for large r (large correlation energy), is the ratio of steady-state correla-

tion energy to (lowest order) steady-state spontaneous- and thermal-emission energy. Comparing Eq. (52) with Eqs. (57) and (59), we see that this ratio is given, for large r, by

$$[8(2\pi^3)^{\frac{1}{2}}/\hbar\omega]NQ_c{}^2u^2\bar{\gamma}^2\langle I\rangle r. \quad (60)$$

One might say, very loosely, that when the absolute value of this ratio is comparable to unity, the behavior of the molecules will become correlated. In that event, higher order interactions than fourth order may also become important.

We consider now some special initial states of the molecules. It is again easier to consider first the situation $T=0$. Then, the only factor in Eq. (44) that depends on the state of the molecules is

$$\langle I\rangle(1+\langle I\rangle), \quad (61)$$

where, it is recalled, $\langle I\rangle$ is the average of $\langle I_m\rangle$, with

$$\langle I_m\rangle=|a_{m2}|^2-|a_{m1}|^2. \quad (62)$$

Considering the dependence of expression (61) on the initial state of the molecules, we see that this expression has a maximum value of 2 when the molecules are all in the upper state; goes down to zero when the molecules are, on the average, in an equal superposition of the upper and lower state; becomes negative as the average expectation value of the molecular energy drops further; but returns to zero when all the molecules are in the lowest energy state. Thus, if one considers the correlation energy as a function of the average energy expectation value, and lets the latter vary from E_2 (upper state) to E_1 (lower state); the correlation energy starts at the maximum; drops to zero when the average expectation value is half-way down; then becomes negative, but returns to zero when the expectation value reaches bottom.

We can understand this behavior, in terms of the picture we have given previously, as an induced effect produced by the lowest order spontaneous emission. When the correlation energy is positive, the induced emission is in phase with the spontaneous emission and reenforces it. When the correlation energy is negative, the induced emission is out of phase with the sponta-neous emission, which means that there is reabsorption of the spontaneous emission, or *radiation trapping*. Going back to the picture of correlated states discussed at the beginning of the article, one can visualize the latter situation as arising from a (correlated) state in which the oscillations of some molecules are out of phase with those of the others.

By setting $T=0$, we have isolated the correlation energy which is due to the spontaneous emission only. The correlation energy produced by thermal effects is the remainder of the correlation energy for $T\neq0$. We see from Eq. (44) that the factor in this thermal part of the correlation energy involving the initial state of the molecules is $\langle I\rangle^2$, so that the thermal correlation

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[9] From a physical, or intuitive, viewpoint, the thermally-induced energy is of a different type than the spontaneous emission energy. The latter may be regarded as being the square of a spontaneous-emission field intensity, and is always positive; the former is the product of the zero-order thermal field intensity and the field intensity which it induces, and can therefore be either positive or negative. The formalism, however, treats these two energies in the same manner; we therefore combine them in order to obtain the complete result in the present argument. For purposes of intuitive clarity, the reader may bear in mind only the sponta-neous-emission energy.

[10] The fact that the intuitive result is somewhat larger than the correct result indicates that the "coherence effects" contained in the higher order processes for the *incoherent* field are not as large as those contained in an independent succession of lower order processes for a *coherent* field. It should also be noted that in the limit of small r we could have obtained, in our intuitive argument, exactly the correct result by using the formula for induced emission due to an incoherent driving field, since in this limit, $R=\frac{1}{2}$. The above formula, however, is entirely inadequate for large r, since $R\rightarrow0$ as $r\rightarrow\infty$. We see, therefore, that the above "coherence effects" vanish when the molecules become nonresonant, but are important for small molecular frequency spread.

energy is always positive. We can understand the origin of this energy in the following manner: The thermal field in the cavity induces oscillation in the molecules. This oscillation of the molecules produces a field the amplitude of which is proportional to the number of molecules, since the molecular oscillations are correlated, having been produced by the same thermal field. The energy, now, is given by the square of the sum of the two field strengths, the zero-order thermal field strength and the induced field strength. The total energy due to thermal effects, therefore contains a mixture term proportional to N[this is the energy of Eq. (54)], which may be positive or negative, depending on the relative phase of the induced field strength[9]; and a square term, which is, of course, always positive and proportional to N^2.

IV

We consider now the energy radiated by the molecules. Formally, this is given by—$\langle \sum_m \dot{H}_m \rangle$. Again, we look for terms which are proportional to N^2 and which are diagonal in the energy representation of the free molecules. We use exactly the same procedure as we did in obtaining the correlation energy of the field, making use of Eqs. (14), (15), and (16). Since the methods of derivation have already been explained and illustrated, we will write down the result only. The correlation power

$$-\langle \sum_m \dot{H}_m{}^{[4]} \rangle = -G[1+f(T)\langle I \rangle]W, \qquad (63)$$

where

$$G = (4\pi^2/\hbar)N^2\omega^3\bar{\gamma}^4u^2\langle I \rangle,$$

and

$$W = \frac{1}{\beta}\left\{\int_0^t d\eta \int_0^{t-\eta} d\eta'\, E(\eta,\eta')e^{-\frac{1}{2}\beta(\eta+\eta')} + \int_0^t d\eta \int_0^\eta d\eta'\, E(\eta,\eta')e^{-\frac{1}{2}\beta(\eta-\eta')}[2+\beta(\eta-\eta')]\right\}$$

$$+\frac{1}{\beta}e^{-\beta t}\int_0^t d\eta \int_{t-\eta}^t d\eta'\, E(\eta,\eta')e^{\frac{1}{2}\beta(\eta+\eta')}.$$

We see that the correlation power radiated by the molecules approaches a steady-state value after a transient period similar to that of the field energy. We also notice, by comparing Eq. (63) with Eqs. (44) and (45), that

$$\beta\langle H_f{}^{[4]} \rangle_{\text{st}} = -\langle \sum_m \dot{H}_m{}^{[4]} \rangle_{\text{st}}, \qquad (64)$$

a very reasonable result, which expresses the fact that in the steady state the correlation power radiated by the molecules is equal to the correlation power absorbed by the loss mechanism of the cavity. In fact, the complete expressions for the radiated power and for the field energy satisfy the relationship

$$\langle \dot{H}_f{}^{[4]} \rangle + \beta\langle H_f{}^{[4]} \rangle = -\langle \sum_m \dot{H}_m{}^{[4]} \rangle, \qquad (65)$$

which states that the power radiated by the molecules is equal to the rate of change of field energy plus power absorbed by the walls.

V

We come now to the last topic under consideration in the present article. So far, we have considered a single cavity mode and a molecular frequency spread centered at the cavity frequency. All other cavity modes were assumed to have frequencies outside the range of the molecular frequency, and therefore do not couple to the molecules. One may ask, however, what the situation would be if there were more than one mode within the molecular frequency range. In order to be able to answer this question by utilizing the results already derived, we introduce simplifying assumptions, which may be somewhat too idealized for a practical situation, but nevertheless lead to a qualitative understanding of the problem involved. We assume that the modes which couple to the molecules all have the same frequency as the single mode previously considered, and that each molecule couples equally to all the modes. Let there be n modes, each labeled with the index ν. Instead of Eq. (4) we have[11]

$$\mathbf{E} = \sum_\nu \mathbf{E}_\nu, \quad \mathbf{H} = \sum_\nu \mathbf{H}_\nu,$$
$$\mathbf{E}_\nu = -4\pi c\mathbf{u}_\nu(\mathbf{r})P_\nu(t), \quad \mathbf{H} = \boldsymbol{\nabla} \times \mathbf{u}_\nu(\mathbf{r})Q_\nu(t), \qquad (66)$$

with

$$[Q_\nu, P_{\nu'}] = i\hbar\delta_{\nu\nu'}.$$

The energy of the field in the cavity is now the sum of the energies of the individual modes:

$$H_f = \sum_\nu [2\pi c^2 P_\nu^2 + (\omega^2/8\pi c^2)Q_\nu^2]. \qquad (67)$$

Our integral equations of motion, Eqs. (14), (15), and (16), become

$$H_m(t) = H_m{}^{[0]} + \frac{2\pi c}{i\hbar}\sum_\nu u_{\nu m}\int_0^t dt_1\{[H_m(t_1), \gamma_m(t_1)], P(t_1)\}, \qquad (68)$$

$$\gamma_m(t) = \gamma_m{}^{[0]}(t) + \frac{4\pi c}{\hbar^2}\sum_\nu u_{\nu m}\int_0^t dt_1 \int_0^{t_1} dt_2 U_m(t-t_1)[\gamma_m(t_1), [\gamma_m(t_2), H_m(t_2)]P(t_2)]U_m{}^{-1}(t-t_1), \qquad (69)$$

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[11] Although the assumption of equal molecular coupling to all the modes is utilized in the final calculations, it is illuminating to write the equations of motion more generally and label each coupling constant $u_{\nu m}$ with the mode index.

and

$$P_\nu = P_{\nu F} + P_{\nu \gamma},\tag{70}$$

where

$$P_{\nu F} = \frac{1}{\omega} \int_{-\infty}^{t} dt_1\, F_\nu(t_1) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1),$$

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

The properties of F_ν are the same as those given by Eqs. (8), (9), and (10) for each individual mode, and we add the condition

$$\langle F_\nu(t_1) F_{\nu'}(t_2)\rangle = 0 \quad \text{for} \quad \nu \neq \nu'.\tag{71}$$

From Eq. (71) we obtain the relationship

$$\langle \{P_{\nu F}(t_1), P_{\nu' F}(t_2)\}\rangle = 0 \quad \text{for} \quad \nu \neq \nu'.\tag{72}$$

We can now proceed exactly as we did in the case of a single mode. In view of the simplifications adopted, we can see that our expressions for the field energy and power radiated by the molecules will be multiples of the corresponding quantities in the case of a single mode, the multiplying factor involving the number of modes. In order to obtain this factor we resort to our shorthand notations and now retain the summations over ν. We consider a term in $\dot{H}_m{}^{[4]}$ that contributes toward the total expression for $\langle \sum_m \dot{H}_m{}^{[4]}\rangle$. From our equations of motion we have

$$\dot{H}_m{}^{[4]} \rightarrow \sum_\nu u_{\nu m} H_m{}^{[3]} \gamma_m{}^{[0]} P_\nu{}^{[0]} + \cdots,\tag{73}$$

$$H_m{}^{[3]} \rightarrow \sum_{\nu'} u_{\nu' m} H_m{}^{[0]} \gamma_m{}^{[0]} P_{\nu'}{}^{[2]} + \cdots,\tag{74}$$

$$P_{\nu'}{}^{[2]} \rightarrow \sum_{m'} \sum_{\nu''} u_{\nu' m'} u_{\nu'' m'} P_{\nu''}{}^{[0]}.\tag{75}$$

Substituting from Eqs. (75) and (74) into Eq. (73), we have

$$\sum_m \dot{H}_m{}^{[4]} \rightarrow \sum_{m,m'} \sum_{\nu,\nu',\nu''} u_{\nu m} u_{\nu' m} u_{\nu' m'} u_{\nu'' m'}$$
$$\times H_m{}^{[0]} \gamma_m{}^{[0]2} P_\nu{}^{[0]} P_{\nu'}{}^{[0]} + \cdots.\tag{76}$$

The right side will lead to a nonvanishing expectation value only for $\nu'' = \nu$. We are thus left with a double summation over ν, which, in view of our assumption of equality of molecular coupling to the various modes, is replaced by n^2. The same reasoning and result apply to the remaining terms in the power radiated by the molecules and to the energy of the field. We see, therefore, that the correlation effects we have been discussing are increased by a factor which is the square of the number of modes coupled to the molecules. It is not difficult to justify this result intuitively. Each molecule is coupled to the others n times as strongly as in the case of a single mode, and also radiates into n modes instead of into one.

ACKNOWLEDGMENT

APPENDIX A

We derive Eq. (34). From Eqs. (9) and (16) we have

$$\langle \{P_F(t), P_F(t')\}\rangle = \frac{\hbar\beta}{2\pi c^2} f(T) \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t'} dt_2\, \delta(t_1-t_2) e^{-\frac{1}{2}\beta(t-t_1)} \sin\omega(t-t_1) e^{-\frac{1}{2}\beta(t'-t_2)} \sin\omega(t'-t_2).\tag{A1}$$

For $t' \geqslant t$,

$$\langle \{P_F(t), P_F(t')\}\rangle = \frac{\hbar\omega\beta}{2\pi c^2} f(T) \int_{-\infty}^{t} dt_1\, e^{-\frac{1}{2}\beta(t+t'-2t_1)} \sin\omega(t-t_1)\sin\omega(t'-t_1) \approx \frac{\hbar\omega}{4\pi c^2} f(T) e^{-\frac{1}{2}\beta(t'-t)} \cos\omega(t'-t),\tag{A2}$$

where we have dropped an oscillatory term in the integrand. For $t > t'$, the integration over t_1 must be carried out before the integration over t_2, and the sign in the exponent of the result in Eq. (A2) is reversed. We have thus derived Eq. (34).

APPENDIX B

We consider the evaluation of

$$X(t) \equiv \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^t dt_4\, EJ,\tag{A3}$$

where

$$E \equiv \exp\{-(1/4\alpha^2)[(t_1-t_2)^2 + (t_3-t_4)^2]\},$$

and

$$J \equiv \exp[-\tfrac{1}{2}\beta(2t-t_1+t_2-t_3-t_4)].$$

Since integration of a Gaussian function over a finite region cannot be carried out explicitly, we alter the variables of integration so that t_1-t_2 and t_3-t_4 are two variables of integration, respectively, and integrate over the other two variables. We therefore set

$$\xi = t_1+t_2, \quad \eta = t_1-t_2,$$
$$\xi' = t_3+t_4, \quad \eta' = t_3-t_4. \tag{A4}$$

Writing the integration symbols as operators, we have

$$X(t) = \frac{1}{4}\int_0^t d\eta \int_\eta^{2t-\eta} d\xi \left\{ \int_0^{\frac{1}{2}(\xi-\eta)} d\eta' \int_{\eta'}^{(\xi-\eta)-\eta'} d\xi' + \int_{\frac{1}{2}(\xi-\eta)}^0 d\eta' \int_{-\eta'}^{(\xi-\eta)-\eta'} d\xi' + \int_{-t}^{\frac{1}{2}(\xi-\eta)-t} d\eta' \int_{-\eta'}^{2t+\eta'} d\xi' \right\} EJ, \tag{A5}$$

where E and J are considered functions of the new variables of integration. We carry out the ξ' integration first. Then we rearrange the order of the ξ and η' integration so that the ξ integration may be carried out next. The result, after some simplification, is given by Eq. (45).

APPENDIX C

We evaluate the expression for X_{st}, in Eq. (46), in the limits of small r and large r, respectively. As its argument approaches zero, the error function vanishes; and as its argument becomes large, we may use the asymptotic formula[12]

$$\text{erf}\,r \approx 1 - [\exp(-r^2)/r\sqrt{\pi}]. \tag{A6}$$

Therefore, in the limit of small r, the first term in the curly bracket of Eq. (46) is equal to π; in the limit of large r, it is equal to r^{-2}. The second term becomes, in the limit of small r,

$$\int_0^\infty dy \int_{-\infty}^\infty dx \, \exp[-\tfrac{1}{8}(x^2+y^2)],$$

which, by transforming to polar coordinates, may easily be seen to be equal to π. In the limit of large r, we have

$$\int_0^\infty dy \int_y^\infty dx \, \exp[-\tfrac{1}{8}(x^2+y^2)](1+ry)e^{-ry} \approx \int_0^\infty dy \, e^{-ry}(1+ry) \int_y^\infty dx \, \exp(-\tfrac{1}{8}x^2)$$

$$= \int_0^\infty dx \, \exp(-\tfrac{1}{8}x^2) \int_0^x e^{-ry}(1+ry)dy \approx \frac{2}{r}\int_{-\infty}^\infty dx \, \exp(-\tfrac{1}{8}x^2) = 2(2\pi)^{\frac{1}{2}}/r. \tag{A7}$$

Combining the above results, we obtain Eqs. (49) and (51).

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[12] H. B. Dwight, *Tables of Integrals* (The Macmillan Company, New York, 1934), Eq. (592).