Field-Correlation Effects in Multiphoton Absorption Processes*

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The dependence of the multiphoton absorption probability for an atomic system on the statistical properties of the incident light is investigated. It is shown, by means of nth-order time-dependent perturbation theory, that the multiphoton absorption probability depends on the (n,n)th normally ordered correlation functions of the electromagnetic field. For stationary fields, the transition probability per unit time is constant and is directly related to the reduced cross-spectral correlation function of the electromagnetic field. This expression for the transition probability is then analyzed under various conditions of coherence. Explicit calculations are carried out for thermal light and for laser light, and the transition rates are compared when each has a Lorentzian spectrum. In particular, when the width of the final atomic level is much larger than the bandwidth of the field, the transition rate for thermal light is $n!$ times higher than with laser light of the same mean intensity. The other extreme case, i.e., when the width of the final atomic level is much smaller than the bandwidth of the field, is also considered in detail. An equation for the density operator of the field is derived; this equation governs the change in the statistical properties of the photon system in n-photon absorption processes. On the basis of this equation, the time-dependent properties of the photon system are then studied and some physical consequences of these equations are discussed. Some numerical solutions relating to the time dependence of the factorial moments in two-photon absorption processes are presented.

I. INTRODUCTION

In the last few years, several papers have appeared which deal with the transition probability for multiphoton processes.[1,2] In these papers, the attention has been mainly focused on the calculation of the transition probability for various such processes. A problem, which is of considerable interest in this connection, concerns the dependence of the transition probability on the statistical nature of the radiation field. Another question of considerable interest is the following one: Do the multiphoton processes provide information about the higher-order correlation functions of the incident electromagnetic field? These two problems, for the particular case of two-photon processes, have been investigated very fully by several workers.[3-12]

The purpose of the present paper is to consider these problems for the case of *multiphoton absorption processes*. We derive, by the use of nth-order time-dependent perturbation theory, an expression for the transition probability for n-photon absorption by an atomic system. The transition probability is found to be dependent on the (n,n)th-order normally ordered correlation function of the electromagnetic field. For absorption with stationary electromagnetic fields, we find that the transition probability per unit time is independent of time and is related to the reduced cross-spectral correlation function of the electromagnetic field. This expression is then analyzed under various conditions of coherence.

Finally, an equation for the density operator of the field is derived. This equation governs the change in the statistical properties of the field in n-photon absorption process and is then used to study the time-dependent statistical properties of the field.

II. MULTIPHOTON-ABSORPTION TRANSITION PROBABILITY

As is well known, the interaction Hamiltonian between the radiation field and an hydrogenlike atom may be written in the form

$$H_I(t) = -\frac{e}{mc}\,\vec{p}\,(t)\cdot\vec{A}(x,t) + \frac{e^2}{2mc^2}\,\vec{A}^{\,2}(x,t). \quad (2.1)$$

Here $\vec{A}(x,t)$ is the vector-potential operator for the electromagnetic field and $\vec{p}(t)$ is the momentum operator of the valence electron. Throughout this paper we shall work in the interaction picture and we shall ignore the effect[13] of the term which is quadratic in $\vec{A}(x,t)$. We shall also use the dipole approximation, i.e., we shall neglect the effect of spatial variation of $\vec{A}(x,t)$. We decompose the vector-potential operator into positive and negative frequency components,

$$\vec{A}(x,t) \simeq \vec{A}(t) = \vec{A}^{(+)}(t) + \vec{A}^{(-)}(t)\,, \quad (2.2)$$

where

$$\vec{A}^{(+)}(t) = \sum_{\vec{k},s}\left(\frac{\hbar c}{kL^3}\right)^{1/2}\vec{\epsilon}(\vec{k},s)\,a_{\vec{k}s}\,e^{-i\omega_k t}\,. \quad (2.3)$$

$\hat{a}_{\vec{k}s}$ in (2.3) is the annihilation operator associated with the mode $(\vec{k},s)$ of the radiation field and

$\epsilon(\vec{k}, s)$ are unit polarization vectors.

The time-development operator in the interaction picture is given by

$$U_I(t) = T\exp[-(i/\hbar)\int_0^t H_I(t')\,dt'] \,, \tag{2.4}$$

where $H_I(t)$ is the interaction Hamiltonian in the interaction picture and T is Dyson's time-ordering operator. We can express (2.4) in the form

$$U_I(t) = \sum_{n=0}^{\infty} U_I^{(n)}(t) \,, \tag{2.5}$$

where
$$\begin{aligned} U_I^{(n)}(t) &= (-i/\hbar)^n(1/n!) \\ &\times \int_0^t \cdots \int T[H_I(t_1)\cdots H_I(t_n)]dt_1\cdots dt_n \\ &= (-i/\hbar)^n \int_0^t \cdots \int \Theta(t_1-t_2)\cdots\Theta(t_{n-1}-t_n) \\ &\times H_I(t_1)\cdot H_I(t_2)\cdots H_I(t_n)dt_1\cdots dt_n \,. \end{aligned} \tag{2.6}$$

Here, $\Theta(\tau)$ is the step function defined by

$$\begin{aligned} \Theta(\tau) &= 1 \,, \quad \text{if} \quad \tau > 0 \\ &= 0 \,, \quad \text{if} \quad \tau < 0 \,. \end{aligned} \tag{2.7}$$

Let $|i\rangle_A$ be the initial state of the atom, assumed to be the ground state, and $|f\rangle_A$ be the final state of the atom. Let $|i\rangle_F$ and $|f\rangle_F$ be the initial and final states of the radiation field, respectively. Then the probability that the system has made a transition to a state $|f\rangle_A \otimes |f\rangle_F$ is given by

$$P(t) = |{}_F\langle f|{}_A\langle f|U_I(t)|i\rangle_A|i\rangle_F|^2 \,. \tag{2.8}$$

Since we are only interested in photon absorption processes, the effective Hamiltonian is

$$H_{\text{eff}}(t) = (-e/mc)\,\vec{p}(t)\cdot\vec{A}^{(+)}(t) \,. \tag{2.9}$$

Since $H_{\text{eff}}(t)$ as given by (2.9), is linear in $\vec{A}^{(+)}(t)$, it is sufficient to replace $U_T(t)$ in (2.8) by $\mathfrak{U}_I^{(n)}(t)$ in order to describe n-photon absorption processes. Here $\mathfrak{U}_I^{(n)}(t)$ is given by

$$\begin{aligned} \mathfrak{U}_I^{(n)}(t) &= (-i/\hbar)n(-e/mc)^n \\ &\times \int_0^t \cdots \int dt_1\cdots dt_n\, \Theta(t_1-t_2)\cdots\Theta(t_{n-1}-t_n) \\ &\times \vec{p}(t_1)\cdot\vec{A}^{(+)}(t_1)\,\vec{p}(t_2)\cdot\vec{A}^{\dagger}(t_2)\cdots\vec{p}(t_n)\cdot\vec{A}^{\dagger}(t_n) \,. \end{aligned} \tag{2.10}$$

Thus the transition probability for n-photon absorption processes is given by

$$P^{(n)}(t) = |{}_F\langle f|{}_A\langle f|\mathfrak{U}_I^{(n)}(t)|i\rangle_A|i\rangle_F|^2 \,. \tag{2.11}$$

For such processes the final state of the field is not known. Hence we sum $P^{(n)}(t)$, given by (2.11), over all the final states of the field. We also suppose that the initial state of the field is described by the density operator

$$\rho_F(0) = \sum_{i_F} \rho_{ii}\,|i\rangle_F\langle i|_F \,. \tag{2.12}$$

Then the expression for the transition probability becomes

$$\begin{aligned} P^{(n)}(t) &= \sum_{i_F f_F} \rho_{ii}\,|\,{}_F\langle f|{}_A\langle f|\mathfrak{U}_I^{(n)}(t)|i\rangle_A|i\rangle_F|^2 \\ &= \sum_{i_F, f_F} \rho_{ii}\;{}_F\langle f|{}_A\langle f|\mathfrak{U}_I^{(n)}(t)|i\rangle_A|i\rangle_F \\ &\quad\times {}_F\langle i|\;{}_A\langle i|\mathfrak{U}_I^{(n)\dagger}(t)|f\rangle_A|f\rangle_F \\ &= \mathrm{Tr}_F\,\rho_F\langle i|U_I^{(n)\dagger}(t)|f\rangle_A\;{}_A\langle f|\mathfrak{U}_I^{(n)}(t)|i\rangle_A \,, \end{aligned} \tag{2.13}$$

where Tr_F denotes the trace over the field variables. Our method follows in part to that employed in Ref. 9. Now following Mollow[9] we introduce the function

$$\Psi_{j_1\cdots j_n}(t_1, t_2, \cdots, t_n) \equiv (e/\hbar mc)^n\,\Theta(t_1-t_2)\cdots\Theta(t_{n-1}-t_n)\,{}_A\langle f|\,p_{j_1}(t_1)\,p_{j_2}(t_2)\cdots p_{j_n}(t_n)|i\rangle_A \,. \tag{2.14}$$

In terms of Ψ, (2.13) can be written in the form

$$\begin{aligned} P^{(n)}(t) &= \sum_{\{i_n\},\{j_n\}} \int_0^t \cdots \int dt_1\cdots dt_n\,dt_1'\cdots dt_n'\,\mathrm{Tr}_F\,\{\rho_F(0)A_{i_1}^{(-)}(t_1')\cdots A_{i_n}^{(-)}(t_n')A_{j_1}^{(+)}(t_1)\cdots A_{j_n}^{(+)}(t_n)\} \\ &\times \Psi^*_{i_1\cdots i_n}(t_1', \cdots, t_n')\,\Psi_{j_1\cdots j_n}(t_1, \cdots, t_n) \,. \end{aligned} \tag{2.15}$$

We shall now express (2.15), in terms of the (n, m)th-order normally ordered correlation function $G^{(n,m)}$ of the electromagnetic field, defined by[14,15]

$$G^{(n,m)}_{i_1\cdots i_n;\,j_1\cdots j_m}(t_1', \cdots, t_n';\, t_1, \cdots, t_m) = \mathrm{Tr}_F\,\{\rho_F(0)A_{i_1}^{(-)}(t_1')\cdots A_{i_n}^{(-)}(t_n')A_{j_1}^{(+)}(t_1)\cdots A_{j_m}^{(+)}(t_m)\} \,. \tag{2.16}$$

Combining (2.15) and (2.16), we obtain the following expression for the transition probability:

$$\begin{aligned} P^{(n)}(t) &= \sum_{\{i_n\}\{j_n\}} \int_0^t \cdots \int dt_1\cdots dt_n\,dt_1'\cdots dt_n'\,G^{(n,n)}_{i_1\cdots i_n;\,j_1\cdots j_n}(t_1', \cdots, t_n';\, t_1, \cdots, t_n) \\ &\times \Psi^*_{i_1\cdots i_n}(t_1', \cdots, t_n')\;\Psi_{j_1\cdots j_n}(t_1, \cdots, t_n) \,. \end{aligned} \tag{2.17}$$

From (2.17) it is clear that the transition probability $P^{(n)}(t)$ depends on the (n,n)th-order normally ordered correlation functions of the electromagnetic field.[16] Hence, experiments on n-photon absorption processes should provide information about correlation functions of the type $G^{(n,n)}$ of the field.[16(a)] It may be shown by straightforward calculation that Ψ is given by

$$\Psi_{\{i_n\}}(\{t_n\}) = [(i)^{n-1}/(2\pi)^{n-1}]\int_{-\infty}^{+\infty}\cdots\int d\{\omega_{n-1}\}\,\Phi_{\{i_n\}}(\{\omega_{n-1}\})\exp[it_1(\omega_f-\omega_1)+it_2(\omega_1-\omega_2)+\cdots+it_n\omega_{n-1}], \tag{2.18}$$

$$\text{where}\quad \Phi_{\{i_n\}}(\{\omega_{n-1}\}) = \left(\frac{e}{\hbar mc}\right)^n \sum_{\{k_{n-1}\}} \frac{{}_A\langle f|p_{i_1}|k_1\rangle_A\cdots{}_A\langle k_{n-1}|p_{i_n}|i\rangle_A}{(\omega_1-\omega_{k_1}+i\epsilon_1)\cdots(\omega_{n-1}-\omega_{k_{n-1}}+i\epsilon_{n-1})}\,. \tag{2.19}$$

Here ϵ_i are infinitesimal positive quantities and the summation in (2.19) is over all the intermediate states of the atom. We have also used the notation $\{\gamma_n\}$ for $\{\gamma_1,\gamma_2,\ldots,\gamma_n\}$. Using (2.17) and (2.18), we obtain

$$P^{(n)}(t) = \int_{-\infty}^{+\infty}\cdots\int d\{\omega'_{n-1};\omega_{n-1}\}\sum_{\{i_n\}\{j_n\}}\Phi^*_{\{i_n\}}(\{\omega'_{n-1}\})\,\Phi_{\{j_n\}}(\{\omega_{n-1}\})$$
$$\times\int_0^t\cdots\int\frac{d\{t'_n;t_n\}}{(2\pi)^{2n-2}}\,G^{(n,n)}_{\{i_n\};\{j_n\}}(\{t'_n\};\{t_n\})\exp[it_1(\omega_f-\omega_1)+\cdots+it_n\omega_{n-1}-it'_1(\omega_f-\omega'_1)-\cdots-it'_n\omega'_{n-1}]. \tag{2.20}$$

For large time intervals ($t\to\infty$), it is evident that the transition probability $P^{(n)}(t)$ tends to $P^{(n)}$ where

$$P^{(n)} = \int_{-\infty}^{+\infty}\cdots\int d\{\omega'_{n-1};\omega_{n-1}\}\sum_{\{i_n\}\{j_n\}}\Phi^*_{\{i_n\}}(\{\omega'_{n-1}\})\,\Phi_{\{j_n\}}(\{\omega_{n-1}\})(2\pi)^2\chi^{(n,n)}_{\{i_n\},\{j_n\}}(\omega_f-\omega'_1,\cdots,$$
$$\omega'_{n-1};\omega_f-\omega_1,\cdots,\omega_{n-1}), \tag{2.21}$$

and where $\chi^{(n,n)}$ is the $2n$th-order cross-spectral correlation function (cf. Ref. 17) and is given by

$$\chi^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega'_n\};\{\omega_n\}) = \frac{1}{(2\pi)^{2n}}\int\cdots\int d\{t'_n;t_n\}\,G^{(n,n)}_{\{i_n\},\{j_n\}}(\{t'_n\};\{t_n\})\exp\left(i\sum_{i=1}^n t_i\omega_i - i\sum_{i=1}^n t'_i\omega'_i\right). \tag{2.22}$$

III. ABSORPTION BY STATIONARY FIELDS

Since practically all the experiments are performed with stationary radiation fields, we shall consider the absorption by stationary fields more fully. A stationary field is one for which the statistical properties are independent of the origin of time, hence for such a field all averages are invariant under time translation. In particular, for stationary fields, one has

$$G^{(n,n)}_{\{i_n\},\{j_n\}}(\{t'_n+\tau\};\{t_n+\tau\}) = G^{(n,n)}_{\{i_n\},\{j_n\}}(\{t'_n\};\{t_n\}), \tag{3.1}$$

where τ is arbitrary time interval. Using a generalization of the Wiener-Khintchine theorem[17] (applied to the present situation), we have, for any stationary field,

$$\chi^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega'_n\};\{\omega_n\}) = \delta\{\sum_{i=1}^n(\omega'_i-\omega_i)\}\,\tilde{\chi}^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega'_n\};\{\omega_{n-1}\},0), \tag{3.2}$$

$$\text{where}\quad \tilde{\chi}^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega'_n\};\{\omega_{n-1}\},0) = \int d\omega_n\,\chi^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega'_n\};\{\omega_n\}) \tag{3.3}$$

$$= \int_{-\infty}^{+\infty}\cdots\int\frac{d\{t'_n;t_{n-1}\}}{(2\pi)^{2n-1}}\,G^{(n,n)}_{\{i_n\},\{j_n\}}(\{t'_n\};\{t_{n-1}\},0)\exp\left(-i\sum_{i=1}^n\omega'_it'_i+i\sum_{i=1}^{n-1}\omega_it_i\right). \tag{3.4}$$

In view of (3.1), the transition probability $P^{(n)}(t)$, as given by (2.20), may also be expressed in the form

$$P^{(n)}(t) = \int_{-\infty}^{+\infty}\cdots\int d\{\omega'_{n-1};\omega_{n-1}\}\sum_{\{i_n\}\{j_n\}}\Phi^*_{\{i_n\}}(\{\omega'_{n-1}\})\,\Phi_{\{j_n\}}(\{\omega_{n-1}\})$$
$$\times\frac{1}{(2\pi)^{2n-2}}\int_0^t dt_n\int_{-t_n}^{t-t_n}d\{t'_n;t_{n-1}\}\exp[-it'_1(\omega_f-\omega'_1)\cdots-it'_n\omega'_{n-1}+it_1(\omega_f-\omega_1)+\cdots$$
$$+it_{n-1}(\omega_{n-2}-\omega_{n-1})]\,G^{(n,n)}_{\{i_n\}\{j_n\}}(\{t'_n\};\{t_{n-1}\},0). \tag{3.5}$$

We now make the usual approximation of the perturbation theory, i.e., we assume that t is much greater than the inverse bandwidth of the field, i.e.,

$$t \gg (1/\Delta\omega). \tag{3.6}$$

We may then extend the limits of the integrals over $\{t'_n;t_{n-1}\}$ to $\pm\infty$ and the transition probability $P^{(n)}(t)$ be-

comes proportional to time. Let $W^{(n)}$ be the transition probability per unit time. According to (3.3), (3.5), and (3.6) it is given by

$$W^{(n)}(\omega_f) = (2\pi) \int_{-\infty}^{+\infty} \cdots \int d\{\omega'_{n-1}; \omega_{n-1}\} \sum_{\{i_n\}\{j_n\}} \Phi_{\{i_n^*\}}(\{\omega'_{n-1}\}) \Phi_{\{j_n\}}(\{\omega_{n-1}\}) \tilde{\chi}_{\{i_n\},\{j_n\}}^{(n,n)}$$

$$\times (\omega_f - \omega'_1, \cdots, \omega'_{n-2} - \omega'_{n-1}, \omega'_{n-1}; \omega_f - \omega_1, \cdots, \omega_{n-2} - \omega_{n-1}, 0) . \tag{3.7}$$

In (3.7) we have displayed explicitly the argument ω_f in order to show the dependence of $W^{(n)}$ on the energy of the final state of the atom. Expression (3.7) shows clearly the dependence of the n-photon absorption probability per unit time on the $2n$th-order reduced cross-spectral (normally ordered) correlation function of the radiation field.

We will now express (3.7) in a slightly different form, which takes into account the symmetry properties of the correlation function $\chi^{(n,n)}$. It is evident that $\chi^{(n,n)}(\{\omega'_n\}; \{\omega_n\})$ is invariant under the permutation of the indices $1, 2, \cdots, n$. In view of this symmetry property, Eq. (3.7) may be rewritten

$$W^{(n)}(\omega_f) = (2\pi) \int_{-\infty}^{+\infty} \cdots \int d\{\omega'_{n-1}; \omega_{n-1}\} \sum_{\{i_n\}\{j_n\}} \tilde{\Phi}_{\{i_n^*\}}(\{\omega'_{n-1}\}) \tilde{\Phi}_{\{j_n\}}(\{\omega_{n-1}\}) \tilde{\chi}_{\{i_n\},\{j_n\}}^{(n,n)}$$

$$\times (\omega_f - \omega'_1, \cdots, \omega'_{n-1}; \omega_f - \omega_1, \cdots, \omega_{n-2} - \omega_{n-1}, 0) , \tag{3.8}$$

where $$\tilde{\Phi}_{\{i_n\}}(\{\omega_{n-1}\}) = (n!)^{-1} [\textstyle\sum_{\Pi} \Phi_{i_{l_1} \cdots i_{l_n}}(\nu_{l_1} \cdots \nu_{l_{n-1}})] . \tag{3.9}$$

The symbol $\sum_{\Pi}$ in (3.9) denotes summation over all the permutation $l_1 \ldots l_n$ of the indices $1, 2 \ldots n$ and the arguments $\nu_{l_1} \cdots \nu_{l_{n-1}}$ are defined by the following set of equations:

$$\omega_f - \nu_{l_1} = \mu_{l_1}, \quad \nu_{l_1} - \nu_{l_2} = \mu_{l_2}, \cdots$$

$$\cdots, \nu_{l_{n-2}} - \nu_{l_{n-1}} = \mu_{l_{n-1}}, \quad \nu_{l_{n-1}} = \mu_{l_n} . \tag{3.10}$$

The quantities $\mu_{l_1} \cdots \mu_{l_n}$ are obtained from the permutation

$$\begin{pmatrix} 1 & 2 \cdots n \\ l_1 & l_2 \cdots l_n \end{pmatrix}$$

of the quantities $\mu_1, \cdots, \mu_n$ defined by the set of relations

$$\omega_f - \omega_1 = \mu_1, \quad \omega_1 - \omega_2 = \mu_2 , \cdots$$

$$\cdots, \omega_{n-2} - \omega_{n-1} = \mu_{n-1}, \quad \omega_{n-1} = \mu_n . \tag{3.11}$$

In particular, for $n = 2$ we have

$$\tilde{\Phi}_{i_1 i_2}(\omega) = \tfrac{1}{2}[\Phi_{i_1 i_2}(\omega) + \Phi_{i_2 i_1}(\omega_f - \omega)] . \tag{3.12}$$

The expression (3.8) will prove very useful in later sections of this paper.

We have so far not taken into account the effects of linewidth of the intermediate and the final atomic states. The effects of the linewidth of the intermediate states are easily taken into account by introducing, in a phenomenological manner, the damping coefficients. Let γ_j be the decay rate due to spontaneous emission from the level j. Then Φ as given by (2.19) is to be replaced by

$$\Phi_{\{i_n\}}(\{\omega_{n-1}\}) = (e/\hbar m c)^n \times \sum_{\{k_{n-1}\}} \frac{{}_A\langle f | p_{i_1} | k_1 \rangle_A \cdots {}_A\langle k_{n-1} | p_{i_n} | i \rangle_A}{(\omega_1 - \omega_{k_1} + i\gamma_1/2) \cdots (\omega_{n-1} - \omega_{k_{n-1}} + i\gamma_{n-1}/2)} . \tag{3.13}$$

In order to take into account the width of the final state, we average the result (3.7), over a line-shape function, which for simplicity we take to be a Lorentzian of width γ_f. The final expression for transition probability is then found to be

$$W^{(n)} = (\gamma_f/2\pi) \int d\omega (\omega^2 + \tfrac{1}{4}\gamma_f^2)^{-1} W^{(n)}(\omega_f + \omega), \tag{3.14}$$

where $W^{(n)}(\omega_f + \omega)$ and Φ are given by (3.7) and (3.13), respectively. In the presence of collisions and also of very intense fields, the Lorentzian line-shape function should be replaced by a more general line-shape function.[18]

A. Absorption by Pure-Mode Fields

We now consider a special class of fields for which there is excitation in only one mode. We

would call such fields pure-mode fields.[19] Such fields are completely coherent in the sense that all even ordered ($m=n$) correlation functions factorize, i.e.,

$$G^{(n,n)}_{\{i_n\}\{j_n\}}(\{t_n'\},\{t_n\})=\prod_{l=1}^{n}[\mathcal{E}^*_{i_l}(t_l')\mathcal{E}_{j_l}(t_l)] \quad . \tag{3.15}$$

Let $\tilde{\mathcal{E}}^{(\omega)}$ be the Fourier transform of $\tilde{\mathcal{E}}(t)$. Then it follows from (3.7) and (3.15) that

$$\begin{aligned}W^{(n)}(\omega_f)=(2\pi)\int_{-\infty}^{+\infty}\cdots\int d\{\omega_{n-1}';\omega_{n-1}\}\\ \times\sum_{\{i_n\}\{j_n\}}\Phi^*_{\{i_n\}}(\{\omega_{n-1}'\})\Phi_{\{j_n\}}(\{\omega_{n-1}\})\\ \times\tilde{\mathcal{E}}^*_{i_1}(\omega_f-\omega_1')\cdots\tilde{\mathcal{E}}^*_{i_n}(\omega_{n-1}')\\ \times\tilde{\mathcal{E}}_{j_1}(\omega_f-\omega_1)\cdots\tilde{\mathcal{E}}_{j_{n-1}}(\omega_{n-2}-\omega_{n-1})\mathcal{E}_{j_n}(0)\,.\end{aligned} \tag{3.16}$$

On the other hand, it is well known[20] that a field which is stationary and mode pure is a monochromatic field, i.e.,

$$\tilde{\mathcal{E}}(\omega)=\delta(\omega-\omega_0)\,\mathcal{E}(\omega_0)\,. \tag{3.17}$$

The final expression for the transition probability with monochromatic fields is obtained by combining (3.16) and (3.17) and the result for *linearly polarized* fields is

$$\begin{aligned}W^{(n)}(\omega_f)=(2\pi)I^n\,\delta(\omega_f-n\omega_0)\\ \times\Phi\{(n-1)\omega_0,\cdots,2\omega_0,\omega_0\}|^2,\end{aligned} \tag{3.18}$$

where I is the intensity of the incident light beam. Thus, one obtains

$$W^{(n)}(\omega_f)\propto I^n\,. \tag{3.19}$$

Hence, the n-photon absorption probability per unit time, for stationary pure-mode fields, is proportional to the nth power of intensity.[21] Note that the δ function in (3.18) corresponds to conservation of energy and in general should be replaced by an appropriate line-shape function. The properties of the atomic system are contained in the quantity $|\Phi|^2$ which may be called the quantum-efficiency parameter of the system (cf. Ref. 16a).

B. Absorption by a Class of Stationary Fields

We now consider an important class of stationary fields for which the phase-space functional[22–24] $F^{(A)}(\{z_{ks}\})$ is dependent only on the moduli of z_{ks} and not on the phases. This class includes thermal field and the field of a single-mode laser.[15] In this case, it can be shown that[17]

$$\langle a^\dagger_{\mu_1'}\cdots a^\dagger_{\mu_n'}a_{\mu_1}\cdots a_{\mu_n}\rangle=\mathcal{S}^{(n)}(\{\mu_n';\mu_n\})\,\chi_n'(\{\mu_n\})\,, \tag{3.20}$$

where, for brevity, we have denoted the indices $(\vec{k},s)$ collectively by the index μ and

$$\mathcal{S}^{(n)}(\{\mu_n';\mu_n\})=\sum_\pi\delta_{\mu_1'\mu_p}\delta_{\mu_2'\mu_q}\cdots\delta_{\mu_n'\mu_r}\,. \tag{3.21}$$

The $\sum_\pi$ in (3.21) denotes summation over all $n!$ permutations of the indices $1,2,\cdots,n$. It can then be shown by straightforward but long calculations that the reduced cross-spectral correlation function is given by

$$\begin{aligned}\tilde{\chi}^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega_n'\};\{\omega_{n-1}\},0)=\left(\frac{\hbar c^2}{L^3}\right)^n\sum_{\{\mu_i'\}\{\mu_i\}}(\omega_{\mu_1'}\cdots\omega_{\mu_n'}\omega_{\mu_1}\cdots\omega_{\mu_n})^{-1/2}\epsilon^*_{i_1}(\mu_1')\cdots\epsilon^*_{i_n}(\mu_n')\\ \times\epsilon_{j_1}(\mu_1)\cdots\epsilon_{j_n}(\mu_n)\mathcal{S}^{(n)}(\{\mu_n';\mu_n\})\left[\prod_{i=1}^{n}\delta(\omega_{\mu_i'}-\omega_i')\prod_{i=1}^{n-1}\delta(\omega_{\mu_i}-\omega_i)\right].\end{aligned} \tag{3.22}$$

On substituting (3.22) in (3.8), we obtain the following expression for the transition probability per unit time:

$$\begin{aligned}W^{(n)}(\omega_f)=(2\pi)(n!)\left(\frac{\hbar c^2}{L^3}\right)^n\sum_{\{\mu_n\}}\chi_n'(\{\mu_n\})(\omega_{\mu_1}\cdots\omega_{\mu_n})^{-1}\,\delta(\omega_f-\sum_{i=1}^{n}\omega_{\mu_i})\\ \times\Big|\sum_{\{j_n\}}\left[\prod_{l=1}^{n}\epsilon_{j_l}(\mu_l)\right]\tilde{\Phi}_{\{j_n\}}\{(\omega_f-\omega_{\mu_1}),\cdots,(\omega_f-\omega_{\mu_1}\cdots-\omega_{\mu_{n-1}})\}\Big|^2\,.\end{aligned} \tag{3.23}$$

In obtaining (3.23) we have also made use of the fact that $\chi_n'(\{\mu_n\})$ is symmetric under the permutation of the indices $1,\cdots,n$ and the definition (3.9).

We again find that, for the particular class of fields considered in this section, the transition probability is completely determined by the reduced cross-spectral density. It seems worth noting that for both thermal field and the laser field oscillating in a single mode, the reduced cross-spectral density factorizes,

i.e.,

$$\chi_n'(\{\mu_i\}) = \prod_{i=1}^{n} \chi_1'(\mu_i'), \tag{3.24}$$

with $\chi_1'(\mu)$ representing the average number of photons in the mode μ.

C. Absorption by Quasimonochromatic Fields near Resonance

Consider a field whose frequency range is confined almost entirely to the domain $\omega_0 - \frac{1}{2}\Delta\omega \leq \omega \leq \omega_0 + \frac{1}{2}\Delta\omega$, where ω_0 is the mean frequency of the field and $\Delta\omega \ll \omega_0$. We shall also assume that the mean frequency ω_0 is near the resonant frequency ω_f/n. Since $\Delta\omega$ is very small, we may ignore the variation of Φ with frequency over the range $|\omega - \omega_0| \leq \frac{1}{2}\Delta\omega$. It should also be noted that the reduced cross-spectral correlation function $\tilde{\chi}^{(n,n)}$ is nonzero only when its arguments lie in the range $|\omega - \omega_0| \leq \frac{1}{2}\Delta\omega$. Hence the expression for the transition probability reduces to

$$W^{(n)}\{\omega_f\} = \sum_{\{i_n\}\{j_n\}} \Phi^*_{\{i_n\}}(\{\omega_0\})\Phi_{\{j_n\}}(\{\omega_0\})\int_{-\infty}^{+\infty} dt\, G^{(n,n)}_{\{i_n\},\{j_n\}}(\{t\};\{0\})\exp[-i\omega_f t], \tag{3.25}$$

where for the sake of brevity, we have denoted the arguments $(\omega_0, \omega_0, \cdots, \omega_0)$ by $\{\omega_0\}$. The result (3.25) is again derived on the assumption that the final state of the atom corresponds to a sharp line with energy $\hbar\omega_f$, which is valid if the lifetime of the final state is much larger than the coherence time of the field. In the general case, we can make use of the result (3.14) to obtain

$$W^{(n)} = \sum_{\{i_n\}\{j_n\}} \Phi^*_{\{i_n\}}(\{\omega_0\})\Phi_{\{j_n\}}(\{\omega_0\})\int_{-\infty}^{+\infty} dt\, G^{(n,n)}_{\{i_n\},\{j_n\}}(\{t\};\{0\})\exp[-i\omega_f t - \tfrac{1}{2}\gamma_f|t|]\ . \tag{3.26}$$

We now define another correlation function $\tilde{G}^{(n,n)}$, which is related to $G^{(n,n)}$ via the following relation:

$$\tilde{G}^{(n,n)}(\{t_n'\};\{t_n\}) \equiv G^{(n,n)}(\{t_n'\};\{t_n\})\exp\left[-i\omega_0\left(\sum_{i=1}^{n}(t_i' - t_i)\right)\right]. \tag{3.27}$$

Here we have separated out the rapidly fluctuating component of the field. On substituting (3.27) in (3.26) and making use of the translation invariance of $G^{(n,n)}$, we obtain the following result:

$$W^{(n)} = 2\sum_{\{i_n\}\{j_n\}} \Phi^*_{\{i_n\}}(\{\omega_0\})\Phi_{\{j_n\}}(\{\omega_0\})\int_{-\infty}^{+\infty} \tilde{G}^{(n,n)}_{\{i_n\},\{j_n\}}(\{-t\};\{t\})\exp[2it(\omega_f - n\omega_0) - \gamma_f|t|]dt\ . \tag{3.28}$$

For linearly polarized fields, (3.28) reduces to

$$W^{(n)} = 2|\Phi(\{\omega_0\})|^2\int_{-\infty}^{+\infty} \tilde{G}^{(n,n)}(\{-t\};\{t\})\exp[2it(\omega_f - n\omega_0) - \gamma_f(t)]dt\ . \tag{3.29}$$

The function $\tilde{G}^{(n,n)}$ is a slowly varying function of its arguments and if the bandwidth of the field is small compared to the width of the final state of the atom ($\Delta\omega \ll \gamma_f$), we may replace $\tilde{G}^{(n,n)}(\{-t\};\{t\})$ in (3.29) by its value for $t=0$. Under these circumstances, (3.29) reduces to

$$W^{(n)} \cong \tilde{G}^{(n,n)}(\{0\};\{0\})|\Phi(\{\omega_0\})|^2\{\gamma_f/[\tfrac{1}{4}\gamma_f^2 + (\omega_f - n\omega_0)^2]\}\ . \tag{3.30}$$

Let us now compute the ratio $W^{(n)}_{\text{th}}/W^{(n)}_{\text{coh}}$, where $W^{(n)}_{\text{th}}$ and $W^{(n)}_{\text{coh}}$ are the transition probabilities for thermal and coherent light, respectively. We have, from (3.30), that

$$W^{(n)}_{\text{th}}/W^{(n)}_{\text{coh}} = G^{(n,n)}_{\text{th}}(\{0\};\{0\})/G^{(n,n)}_{\text{coh}}(\{0\};\{0\})\ . \tag{3.31}$$

For thermal field one has[25]

$$G^{(n,n)}_{\text{th}}(\{0\};\{0\}) = n!\,\langle I\rangle^n \tag{3.32}$$

and for coherent field

$$G^{(n,n)}_{\text{coh}}(\{0\};\{0\}) = \langle I\rangle^n\ . \tag{3.33}$$

For the purpose of comparison, we assume that both the fields have the same mean intensity $\langle I\rangle$ and then from (3.31)–(3.33) it follows that

$$W^{(n)}_{\text{th}}/W^{(n)}_{\text{coh}} = n!\ . \tag{3.34}$$

Thus we conclude that for quasimonochromatic fields near resonance and with $\gamma_f \gg \Delta\omega$, the transition probability with thermal light is $n!$ times higher than with coherent light.[26]

D. n-Photon Absorption with Thermal Light

For a field in thermal equilibrium it is well known[15] that the higher-order correlation functions may be

expressed in terms of second-order correlation functions by means of the relation

$$G^{(n,n)}_{\{i_n\},\{j_n\}}(\{t'_n\};\{t_n\}) = \sum_\pi G^{(1,1)}_{i_1 j_{l_1}}(t'_1, t_{l_1}) \cdots G^{(1,1)}_{i_n, j_{l_n}}(t'_n, t_{l_n}) \ , \tag{3.35}$$

and $G^{(n,m)}_{\{i_n\},\{j_m\}}(\{t'_n\};\{t_m\}) = 0 \ , \quad n \neq m$ (3.36)

where $\sum_\pi$ stands for the sum over all $n!$ possible permutations of the indices 1 to n. Since the field is also stationary $G^{(1,1)}_{i,j}(t';t)$ depends only on the time difference $(t-t')$. Let

$$G^{(1,1)}_{i,j}(t';t) \equiv \Gamma_{ij}(t-t') \ . \tag{3.37}$$

The correlation function $\Gamma_{ij}(\tau)$ is easily calculated[15] from the properties of the thermal field. Let $s(\omega)$ be the spectrum of the field; then by Wiener-Khintchine theorem

$$s(\omega) = \int_{-\infty}^{+\infty} dt \ \Gamma(\tau) e^{i\omega\tau} \ . \tag{3.38}$$

From (3.35), (3.38), and (2.22), it may be shown that

$$\chi^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega'_n\};\{\omega_n\}) = \frac{1}{(2\pi)^n} \sum_\pi \delta(\omega'_1 - \omega_{l_1}) \cdots \delta(\omega'_n - \omega_{l_n}) s_{i_1 j_{l_1}}(\omega'_1) \cdots s_{i_n j_{l_n}}(\omega'_n) \ . \tag{3.39}$$

Using (3.39), we may easily calculate the reduced cross-spectral correlation function $\tilde{\chi}^{(n,n)}$, and we find that

$$\tilde{\chi}^{(n,n)}_{\{i_n\},\{j_n\}}(\{\omega'_n\};\{\omega'_{n-1}\},0) = \frac{1}{(2\pi)^n} \sum_\pi s_{i_1 j_{l_1}}(\omega'_1) \cdots s_{i_n j_{l_n}}(\omega'_n) \prod_{\substack{j=1 \\ (l_j \neq n)}}^{n} \delta(\omega'_j - \omega_{l_j}) \ . \tag{3.40}$$

The final expression for the transition probability would be obtained by substituting (3.40) in (3.8). In view of the definition of $\tilde{\Phi}$, it is easily seen that each term under the summation sign would give the same contribution to W^n. Hence the transition probability per unit time for n-photon absorption with thermal light is given by

$$W^{(n)}_{\text{th}}(\omega_f) = (2\pi) n! \frac{1}{(2\pi)^n} \int_{-\infty}^{+\infty} \cdots \int d\{\omega_{n-1}\} \sum_{\{i_n\}\{j_n\}} \tilde{\Phi}^*_{\{i_n\}}(\{\omega_{n-1}\}) \tilde{\Phi}_{\{j_n\}}(\{\omega_{n-1}\}) s_{i_1 j_1}(\omega_f - \omega_1) \cdots s_{i_n j_n}(\omega_{n-1}) . \tag{3.41}$$

This is our final expression for n-photon absorption transition probability with thermal fields.

We again consider the case of a narrow-bandwidth field near resonance and assume that the field is linearly polarized. Then (3.29) gives

$$W^{(n)} = 2|\Phi(\{\omega_0\})|^2 \int_{-\infty}^{+\infty} dt \ G^{(n,n)}(\{-t\};\{t\}) \exp[2it\,\omega_f - \gamma_f |t|] \ dt \ . \tag{3.42}$$

Use of (3.35) leads to the following relation for thermal fields:

$$G^{(n,n)}(\{-t\};\{t\}) = n! \, [\Gamma(2t)]^n . \tag{3.43}$$

On substituting (3.43) in (3.42), we obtain

$$W^{(n)} = n! \ |\Phi(\{\omega_0\})|^2 \int_{-\infty}^{+\infty} dt [\Gamma(t)]^n \ \exp[it\,\omega_f - \tfrac{1}{2}\gamma_f |t|] \ . \tag{3.44}$$

We now evaluate the integral appearing in (3.44) for the case when the spectrum of the incident light is Lorentzian, i.e., when

$$\Gamma(t) = \langle I \rangle \exp[-i\omega_0 t - b|t|] \ , \tag{3.45}$$

where $\langle I \rangle$ is the mean intensity of the light beam and b is the half-width of the spectrum. From (3.44) and (3.45), we may easily show that

$$W^{(n)} = \langle I \rangle^n |\Phi(\{\omega_0\})|^2 n! (2nb + \gamma_f) / [(nb + \tfrac{1}{2}\gamma_f)^2 + (\omega_f - n\omega_1)^2] \ . \tag{3.46}$$

E. Absorption with Well-Stabilized Laser Beam

In this section, we consider the absorption rate with a well-stabilized laser beam. We assume the phase diffusion model[27] for the laser beam, i.e., we assume that the field amplitude is constant but the frequency is a random variable, to be described shortly. Our analysis will be based on the theory of stochastic processes. More precisely we assume that the complex amplitude of the field (considered to be linearly

polarized) may be represented in the form

$$V(t)=I^{1/2}\exp[-i\int_0^t\omega(t')dt'-i\varphi_0], \tag{3.47}$$

where $V(t)$ is a random variable. In (3.47), I is the intensity of the light beam and φ_0 is a random variable which is uniformly distributed between 0 and 2π, to preserve stationarity. Moreover $\omega(t')$ is assumed to be

$$\omega(t')=\omega_0+\Delta\omega(t)\ , \tag{3.48}$$

where ω_0 is the mean frequency and $\Delta\omega(t)$ is a slowly varying function of time. We assume that the statistical properties of $\Delta\omega(t)$ are those of a δ-*correlated real Gaussian random process*. This shows that the autocorrelation function has the form

$$\langle\Delta\omega(t')\ \Delta\omega(t'')\rangle=2b\,\delta(t'-t'') \tag{3.49}$$

and that the characteristic functional $C[f(\circ)]$, i.e.,

$$C[f(\circ)]=\langle\exp[i\int dt\ \Delta\omega(t)f(t)]\rangle, \tag{3.50}$$

has the form[28]

$$C[f(\circ)]=\exp[-\tfrac{1}{2}\int\int f(t')f(t'')\langle\Delta\omega(t')\ \Delta\omega(t'')\rangle dt'dt'']. \tag{3.51}$$

Using (3.47), (3.48), (3.49), and (3.51) it can be shown that

$$G^{(n,n)}(\{t_n'\};\{t_n\})\equiv\langle V^*(t_1')\cdots V^*(t_n')V(t_1)\cdots V(t_n)\rangle$$

$$=I^n\exp\left[i\omega_0\left(\sum_{i=1}^{n}{}'(t_i'-t_i)\right)\right]\exp\left[b\left(\sum_{\substack{ij\\ i<j}}(|t_i'-t_j'|+|t_i-t_j|)-\sum_{ij}|t_i'-t_j|\right)\right]. \tag{3.52}$$

Hence for this model one has

$$G^{(n,n)}(\{-t\};\{t\})=I^n\exp[-2in\omega_0t-2bn^2|t|]\ . \tag{3.53}$$

Let us assume that the bandwidth of the field is so small that the approximations made in Sec. III C are valid. On combining (3.29) and (3.53), we obtain the following expression for the expression for the transition probability for n-photon absorption with laser light:

$$W_{laser}^{(n)}=I^n|\Phi(\{\omega_0\})|^2(\gamma_f+2bn^2)/[(\tfrac{1}{2}\gamma_f+bn^2)^2+(\omega_f-n\omega_0)^2]. \tag{3.54}$$

It is of interest to compare the transition rate $W_{\rm th}^{(n)}$ and $W_{\rm laser}^{(n)}$ as given by (3.46) and (3.54), respectively. We have already considered in Sec. III C the case when $\gamma_f\gg b$. We now discuss the other extreme $\gamma_f\ll b$, i.e., when the width of the final state is much smaller than the bandwidth of the field. In this case, (3.46) and (3.54) reduce to

$$W_{\rm th}^{(n)}=|\Phi(\{\omega_0\})|^2\langle I\rangle^n(2)n!(bn)/[(nb)^2+(\omega_f-n\omega_0)^2], \tag{3.55}$$

$$W_{\rm laser}^{(n)}=I^n|\Phi(\{\omega_0\})|^2(2)(bn^2)/[(bn^2)^2+|\omega_f-n\omega_1)^2]. \tag{3.56}$$

When the field is near the resonance, i.e., when $\omega_f\approx n\omega_0$, one finds from (3.55) and (3.56) that

$$W_{\rm th}^{(n)}/W_{\rm laser}^{(n)}=(n)(n!),\quad\text{for}\quad\gamma_f\ll b,\quad(n\omega_0-\omega_f)\ll nb. \tag{3.57}$$

Here for the sake of comparison we assumed that the two light beams have the same mean intensities. If the field is far off the resonance but still satisfying the conditions of Sec. III C, then the ratio reduces to

$$W_{\rm th}^{(n)}/W_{\rm laser}^{(n)}=(n-1)!,\quad\text{for}\quad\gamma_f\ll b,\quad(n\omega_0-\omega_f)\gg nb. \tag{3.58}$$

It is worth mentioning that all these results reduce to Mollow's results[9] on two-photon absorption process, if one substitutes $n=2$ in Eqs. (3.54)–(3.58). Similar calculations can be carried out for the laser oscillating in two modes.

4. EQUATION OF MOTION FOR ρ_f

Having studied the effects of the coherence properties of the incident electromagnetic field on the multiphoton absorption probability, we proceed to investigate the changes in the statistical properties of the radiation field itself as the photon absorption process goes on. In other words, we

shall first obtain an equation of motion for the density operator of the field alone. The problem is much more complex than the problem considered in earlier sections. To do this, we will follow a slightly different approach. Let $|1\rangle$ and $|2\rangle$ be the states of the atom which take part in the n-photon absorption processes, i.e., the atom makes transition from the state $|1\rangle$ to the state $|2\rangle$ by absorbing one photon from, say, each of the modes $k_1, k_2, \cdots, k_n$. We assume that the only resonance which exists corresponds to $\omega_{12} = \omega_{k_1} + \cdots + \omega_{k_n}$, where ω_{12} is the energy separation between the levels $|1\rangle$ and $|2\rangle$. Following Shen[6] we may express the interaction Hamiltonian for this process in the form

$$H_{\text{int}} = \sum_i \{\eta c_{2i}^\dagger c_{1i} E_{k_1}^\dagger(\vec{r}_i) \cdots E_{k_n}^\dagger(\vec{r}_i) + \text{H.c.}\} . \tag{4.1}$$

Here c_{1i}, c_{2i}, $c_{1i}^\dagger$, and $c_{2i}^\dagger$ are the annihilation and creation operators for the ith atom in states $|1\rangle$ and $|2\rangle$, respectively. η is the matrix element[29] for n-photon absorption processes. The positive-frequency part of the electric field at the ith atom is given by

$$E_k^\dagger(\vec{r}_i) = (-i)(2\pi\hbar\omega_k)^{1/2} u_k(\vec{r}_i) a_k, \tag{4.2}$$

where $u_k(\vec{r}_i)$ are the mode eigenfunctions. It should also be noted that (4.1) will, in general, involve a summation over all those modes whose frequencies sum to ω_{12}. For the sake of simplicity we assume that $k_1, k_2, \cdots, k_n$ are the only modes whose frequencies sum to ω_{12}. However, the generalization of this to the other case is trivial. The density operator in the interaction picture satisfies the following equation of motion:

$$i\hbar \frac{\partial \rho}{\partial t} = [H_I(t), \rho(t)], \tag{4.3}$$

where $H_I(t)$ is the interaction Hamiltonian in the interaction picture. Note that the Hamiltonian given by (4.1) is of order n in the creation and the annihilation operators and therefore to describe n-photon absorption processes, it is sufficient to carry out the perturbation calculations on Eq. (4.3) to the second order. This is one of the advantages of expressing the Hamiltonian in the form (4.1). Of course one can also start with the Hamiltonian (2.1) and perform $2n$th-order perturbation on Eq. (4.3). However, this latter method becomes extremely cumbersome and for this reason we adopt the first method.

We assume that at some initial time the atomic system is in the thermal equilibrium at temperature T. The density operator ρ_F of the field alone at time t is given by

$$\rho_F(t) = \text{Tr}_A\{\rho(t)\}, \tag{4.4}$$

where Tr_A denotes the trace operation over the atomic variables. We also assume that at time $t = 0$, when the interaction is switched on, the radiation field and the atomic system are decoupled, i.e., that

$$\rho(0) = \rho_F(0) \otimes \rho_A(0) , \tag{4.5}$$

$$\rho_A(0) = \Pi_i \rho_i(0) , \tag{4.6}$$

where $\rho_i(0)$ is the thermal-equilibrium density operator for the ith atom. Thus, the equation of motion for ρ_F can be obtained from (4.1) and (4.3)–(4.5). Standard second-order perturbation procedure[30] leads to

$$\begin{aligned}\frac{\partial \rho_F}{\partial t} = &-\lambda^{(n)}\kappa_1\{a_{\kappa_n}^\dagger \cdots a_{\kappa_1}^\dagger a_{\kappa_1} \cdots a_{\kappa_n}\rho_F - 2a_{\kappa_1} \cdots a_{\kappa_n}\rho_F a_{\kappa_n}^\dagger \cdots a_{\kappa_1}^\dagger + \rho_F a_{\kappa_n}^\dagger \cdots a_{\kappa_1}^\dagger a_{\kappa_1} \cdots a_{\kappa_n}\} \\ &-\lambda^{(n)}\kappa_2\{a_{\kappa_1} \cdots a_{\kappa_n} a_{\kappa_n}^\dagger \cdots a_{\kappa_1}^\dagger \rho_F - 2a_{\kappa_n}^\dagger \cdots a_{\kappa_1}^\dagger \rho_F a_{\kappa_1} \cdots a_{\kappa_n} + \rho_F a_{\kappa_1} \cdots a_{\kappa_n} a_{\kappa_n} \cdots a_{\kappa_1}^\dagger\} ,\end{aligned} \tag{4.7}$$

where κ_1 and κ_2 are the thermal populations of the two atomic states $|1\rangle$ and $|2\rangle$. $\lambda^{(n)}$ is the absorption coefficient for n-photon absorption process and is given by

$$\lambda^{(n)} = (2\pi\hbar)^{n-2}(2\pi^2)(\omega_{\kappa_1} \cdots \omega_{\kappa_n})|\eta|^2 g(\omega_{\kappa_1} + \cdots + \omega_{\kappa_n}) \int d^3r N(\vec{r}) |u_{\kappa_1}(\vec{r})|^2 \cdots |u_{\kappa_n}(\vec{r})|^2 . \tag{4.8}$$

Here $g(\omega)$ is the line-shape function and $N(\vec{r})$ is the density of atoms at the position $\vec{r}$ in the cavity. The integration in (4.8) extends over the whole volume of the medium.

We shall from now on consider the case when all the incident field is mode pure. Then Eq. (4.7) reduces to

$$\frac{\partial \rho_F}{\partial t} = -\lambda^{(n)}\kappa_1(a^{\dagger n}a^n\rho_F - 2a^n\rho_F a^{\dagger n} + \rho_F a^{\dagger n}a^n) - \lambda^{(n)}\kappa_2(a^n a^{\dagger n}\rho_F - 2a^{\dagger n}\rho_F a^n + \rho_F a^n a^{\dagger n}) , \tag{4.9}$$

where for the sake of brevity we have not indicated a mode label here. Equation (4.9) is the desired equation for and will be used to discuss the statistical properties of the field mode in n-photon pro-

cesses. In particular from (4.9), it is easy to show that

$$\frac{\partial\langle a\rangle_t}{\partial t} = -\lambda^{(n)}\kappa_1 n\langle a^{\dagger n-1}a^n\rangle_t + \lambda^{(n)}\kappa_2 n\langle a^n a^{\dagger n-1}\rangle_t \ , \quad (4.10)$$

$$\frac{\partial\langle a^\dagger a\rangle_t}{\partial t} = -2n\lambda^{(n)}\kappa_1\langle a^{\dagger n}a^n\rangle_t + 2n\lambda^{(n)}\kappa_2\langle a^n a^{\dagger n}\rangle_t \ , \quad (4.11)$$

where quantities such as $\langle f(a,a^\dagger)\rangle_t$ represent the average value at time t, i.e.,

$$\langle f(a,a^\dagger)\rangle_t = \mathrm{Tr}\{\rho_F(t)f(a,a^\dagger)\} \ . \quad (4.12)$$

It is to be noted that Eq. (4.11) shows some interesting features of the coupling. The first term in (4.11) is proportional to the (n,n)th normally ordered correlation function of the field amplitude, whereas the second term is proportional to the (n,n)th antinormally ordered correlation function of the field amplitude. These two terms correspond to pure n-photon absorption and pure n-photon emission, respectively. At zero temperature ($\kappa_1=1$, $\kappa_2=0$), Eq. (4.11) reduces to

$$\frac{\partial\langle a^\dagger a\rangle_t}{\partial t} = -2n\lambda^{(n)}\langle a^{\dagger n}a^n\rangle_t \ . \quad (4.13)$$

It is clear from (4.13) that the rate of change of the average numbers of photons in the field depends on the statistical nature of the light. We also know that

$$\langle a^{\dagger n}a^n\rangle_{\mathrm{th}} = n!\langle a^\dagger a\rangle_{\mathrm{th}}^n \ , \quad (4.14)$$

$$\langle a^{\dagger n}a^n\rangle_{\mathrm{coh}} = \langle a^\dagger a\rangle_{\mathrm{coh}}^n \ . \quad (4.15)$$

It therefore follows from (4.13)–(4.15), that the absorption rate with thermal light would be $n!$ times higher than with coherent light.

Another important feature of (4.11) should be noted. It is obvious that the first moment of the number operator is coupled to the nth normally ordered and antinormally ordered moments of the number operator. These moments in turn are coupled to higher-order moments. One in this way obtains an infinite hierarchy of equations. In the later part of this section, we shall give a method of solving this infinite hierarchy and carry out some numerical computations for the case of two-photon absorption processes.

It is also of interest to obtain equation for the diagonal matrix elements of the density operator $\rho_F(t)$. Let $\rho_m(t)$ equal

$$p_m(t) = \langle m|\rho_f(t)|m\rangle \ , \quad (4.16)$$

where $|m\rangle$ denotes the Fock state of the radiation field. Obviously $p_m(t)$ is the probability that there are m photons in the field at time t. From (4.9) and (4.16), we obtain

$$\begin{aligned}\frac{\partial p_m(t)}{\partial t} &= -2\lambda^{(n)}\kappa_1\left[\frac{m!}{(m-n)!}\right]p_m(t) \\ &\quad -2\lambda^{(n)}\kappa_2\left[\frac{(m+n)!}{m!}\right]p_m(t) + 2\lambda^{(n)}\kappa_1 \\ &\quad \times\left[\frac{(m+n)!}{m!}\right]p_{m+n}(t) + 2\lambda^{(n)}\kappa_2 \\ &\quad \times\left[\frac{m!}{(m-n)!}\right]p_{m-n}(t) \ . \end{aligned} \quad (4.17)$$

Again (4.17) represents an infinite hierarchy of equations. In case when the absorption processes are neglected and one considers the emission processes alone, one can solve (4.17) by a set of recurrence relations. For the case of two-photon emission processes, the solutions have been given by Lambropoulos.[11] In order to decouple this hierarchy of equations, we introduce the generating function $Q(x,t)$, for $p_m(t)$, defined as

$$Q(x,t) = \sum_{n=0}^{\infty}(1-x)^n p_n(t) \ . \quad (4.18)$$

From (4.18) and (4.17), one can show by straightforward but long calculation, that the generating function $Q(x,t)$ satisfies the following nth-order partial differential equation:

$$\begin{aligned}\frac{\partial Q(x,t)}{\partial t} &= -2\lambda^{(n)}\kappa_1(-1)^n\left[(1-x)^n\frac{\partial^n Q}{\partial x^n}\right] \\ &\quad +2\lambda^{(n)}\kappa_1(-1)^n\frac{\partial^n Q}{\partial x^n} - 2\lambda^{(n)}\kappa_2(-1)^n\frac{\partial^n}{\partial x^n}\{(1-x)^n Q\} \\ &\quad +2\lambda^{(n)}\kappa_2(-1)^n(1-x)^n\frac{\partial^n}{\partial x^n}[(1-x)^n Q] \ . \end{aligned} \quad (4.19)$$

Equation (4.19) has to be solved for the generating function $Q(x,t)$ subject to appropriate initial condition. At *zero temperature*, and for two-photon absorption processes, (4.19) reduces to

$$\frac{\partial Q}{\partial t} = 2\lambda^{(2)}(2x-x^2)\frac{\partial^2 Q}{\partial x^2} \ . \quad (4.20)$$

Let us introduce a parameter τ defined by

$$\tau = 2\lambda^{(2)}t \ . \quad (4.21)$$

Then (4.20) becomes

$$\frac{\partial Q}{\partial \tau} = (2x-x^2)\frac{\partial^2 Q}{\partial x^2} \ . \quad (4.22)$$

We shall first find an exact solution of (4.22). Let us set

$$y = (1-x), \qquad Q(x,\tau) \equiv R(y,\tau) \ . \quad (4.23)$$

Then, from (4.22), we obtain

$$\frac{\partial R}{\partial \tau} = (1-y^2)\frac{\partial^2 R}{\partial y^2} \ . \quad (4.24)$$

Equation (4.24) can be solved in terms of ultra spherical polynomials[31] $C_n^{(-1/2)}(y)$ and the solution is given by

$$R(y,\tau)=\sum_0^\infty d_n C_n^{(-1/2)}(y)e^{-n(n-1)\tau} \quad . \tag{4.25}$$

The coefficients d_n have to be determined from the initial value $R(y,0)$. Some of the more important properties of these polynomials are given by the following equations:

$$(1-2yz+z^2)^{1/2}=\sum_0^\infty C_n^{(-1/2)}(y)z^n, \quad (|z|<1) \tag{4.26}$$

$$\frac{d}{dy}C_n^{(-1/2)}(y)=-P_{n-1}(y), \qquad (n\geq 1) \tag{4.27a}$$

$$\frac{d}{dy}C_0^{(-1/2)}(y)=0 \quad , \tag{4.27b}$$

$$\int_{-1}^{+1}[dy/(1-y^2)]C_n^{(-1/2)}(y)C_{n'}^{(-1/2)}(y) = [n(n-\tfrac{1}{2})(n-1)]^{-1/2}\delta_{nn'}, \qquad (n\geq 2) \quad . \tag{4.28}$$

If a function $f(y)$ satisfies the same boundary conditions as $C_n^{(-1/2)}(y)$, then the following expansion holds:

$$f(y)=\sum_0^\infty d_n C_n^{(-1/2)}(y) \quad , \tag{4.29}$$

where $d_0=\frac{1}{2}[f(-1)+f(1)]$, $d_1=\frac{1}{2}[f(-1)-f(1)]$,

$$d_n=n(n-1)(n-\tfrac{1}{2}) \tag{4.30}$$

$$\times\int_{-1}^{+1}f(y)C_n^{(-1/2)}(y)[dy/(1-y^2)], \quad (n\geq 2) \quad . \tag{4.31}$$

Returning to (4.25), we find that the coefficients d_n are given by

$$\begin{aligned} d_0&=\tfrac{1}{2}[R(-1,0)+R(1,0)] \ , \\ d_1&=\tfrac{1}{2}[R(-1,0)-R(1,0)] \quad . \\ d_n&=n(n-1)(n-\tfrac{1}{2}) \\ &\quad\times\int_{-1}^{+1}R(y,0)\,C_n^{(-1/2)}(y)[dy/(1-y^2)], \quad (n>1). \end{aligned} \tag{4.32, 4.33}$$

Once the solution of (4.25) is known we can use it to discuss the statistical properties of the photon system in two-photon absorption processes. For example, the average number of photons $\langle n(\tau)\rangle$ at time τ is given by

$$\langle n(\tau)\rangle = -\frac{\partial Q(x\tau)}{\partial x}\bigg|_{x=0}=\frac{\partial R(y,\tau)}{\partial y}\bigg|_{y=1} \quad . \tag{4.34}$$

From (4.34) and (4.25) and (4.27a), we find that

$$\langle n(\tau)\rangle = \sum_0^\infty d_n e^{-n(n-1)\tau}\frac{\partial C_n^{(-1/2)}(y)}{\partial y}\bigg|_{y=1}$$

$$=-\sum_1^\infty d_n e^{-n(n-1)\tau}\,P_{n-1}(1) \ ,$$

$$\text{or} \quad \langle n(\tau)\rangle=-\sum_1^\infty d_n\exp[-n(n-1)\tau] \ , \tag{4.35}$$

where the coefficients d_n are given by (4.32) and (4.33).

Having obtained the exact solution, we will now present some numerical results. We assume that at time $t=0$, the probability $p_n(0)$ that the system is in Fock state $|n\rangle$ is given by Poisson distribution, i.e.,

$$p_m(0)=e^{-\bar{n}}(\bar{n}^m/m!) \ , \tag{4.36}$$

where $\bar{n}$ is the average number of photons in the system at time $t=0$. From (4.18) and (4.36), we find that

$$Q(x,0)=e^{-\bar{n}x} \ . \tag{4.37}$$

We have carried out the numerical computations for the case when $\bar{n}$ was equal to 10. The differential equation (4.22) was solved numerically and the following quantities were computed:

$$\langle n(t)\rangle \equiv \langle a^\dagger a\rangle_t=-\frac{\partial Q(x,t)}{\partial x}\bigg|_{x=0} \ , \tag{4.38}$$

$$\langle n^{[2]}(t)\rangle \equiv \langle a^\dagger a\,a^\dagger a\rangle_t-\langle a^\dagger a\rangle_t=\frac{\partial^2 Q(x,t)}{\partial x^2}\bigg|_{x=0} \ , \tag{4.39}$$

$$\begin{aligned}\langle[\Delta n(t)]^2\rangle &\equiv \langle n^2(t)\rangle-\langle n(t)\rangle^2 \\ &=\langle n^{[2]}(t)\rangle+\langle n(t)\rangle-2n(t)\rangle^2 \ .\end{aligned} \tag{4.40}$$

The results of these numerical computations[32] are shown in Figs. 1–3, respectively. It is seen from the curves that as the time increases, the first and second factorial moments of n, as well as the variance, decrease continuously as expected. For small times the depletion is quite rapid. For large time $\tau(\sim 1)$, steady state is reached. The behavior of the curve of Fig. 1 can also be understood by examining the equation of motion for $\langle n(t)\rangle$. It is easily seen from (4.22) that $\langle n(t)\rangle$ satisfies the equation

$$\frac{\partial\langle n(\tau)\rangle}{\partial\tau}=-2\{\langle n^2(\tau)\rangle-\langle n(\tau)\rangle\} \ . \tag{4.41}$$

We note that the operator $\{n^2-n\}\equiv\{(a^\dagger a)^2-(a^\dagger a)\}$ is a positive-definite operator and hence its expectation value in any state should satisfy the following relation:

$$\begin{aligned}&\mathrm{Tr}[\rho_F(0)\{(a^\dagger(\tau)a(\tau))^2-a^\dagger(\tau)a(\tau)\}] \\ &=\mathrm{Tr}[\rho_F(\tau)\{(a^\dagger a)^2-(a^\dagger a)\}]\geq 0 \ .\end{aligned} \tag{4.42}$$

Since $\rho_F(t)$ is also a positive-definite operator, we conclude that

$$\langle n^{[2]}(t)\rangle \geq 0 \ . \tag{4.43}$$

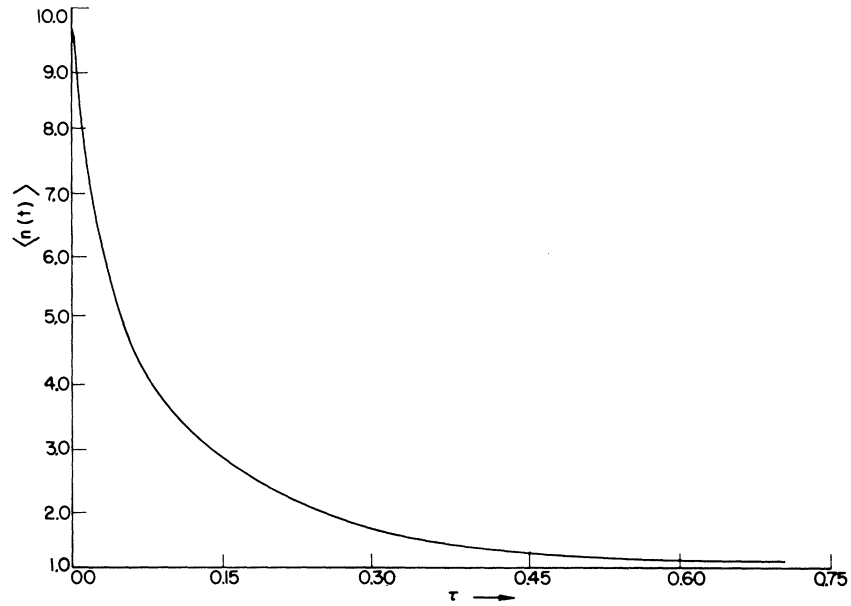

FIG. 1. Average number of photons $\langle n(t)\rangle$ as a function of $\tau = 2\lambda^{(2)}t$.

From (4.41) and (4.43), we have

$$\frac{\partial\langle n(\tau)\rangle}{\partial\tau} \leq 0, \quad \text{for all} \quad \tau . \tag{4.44}$$

Equation (4.44) is the statement that $\langle n(\tau)\rangle$ should decrease as time increases. On comparing curves 1 and 3, we see that

$$\langle n(\tau)\rangle \simeq \langle[\Delta n(\tau)]^2\rangle . \tag{4.45}$$

Equation (4.45) should be compared with the relation which applies to Poisson distribution,

$$\langle n(\tau)\rangle = \langle[\Delta n(\tau)]^2\rangle . \tag{4.46}$$

Equations of the type (4.41) have also been encountered in connection with studies on the time-dependent statistical properties of the laser radiation.[33] One can carry out the similar numerical computations, for the case when $p_m(0)$ is given by the Bose-Einstein distribution, appropriate to thermal light. As predicted by general principles in earlier sections, one would find that the depletion is twice as rapid as in case when $p_m(0)$ is given by the Poisson distribution.

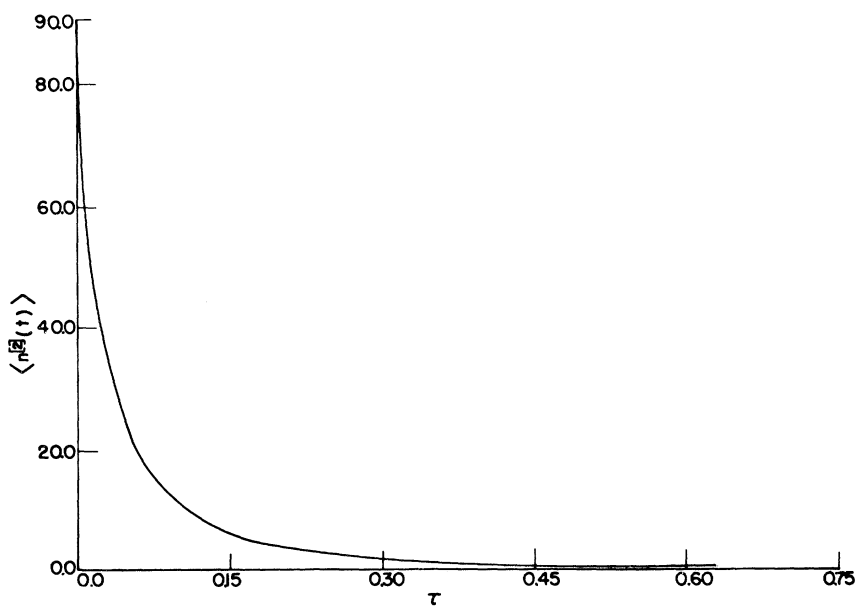

FIG. 2. Second factorial moment $\langle n^{[2]}(t)\rangle$ as a function of $\tau = 2\lambda^{(2)}t$.

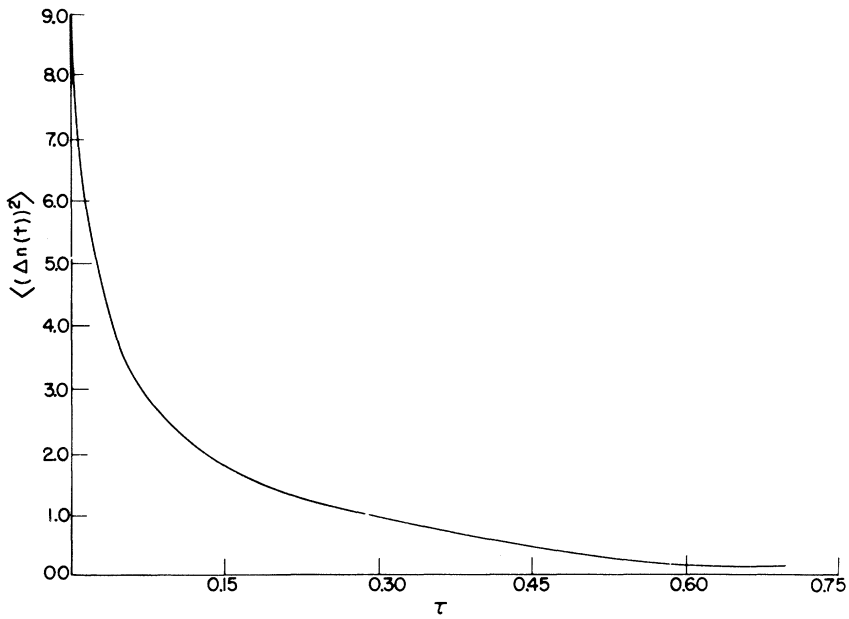

FIG. 3. Variance $\langle[\Delta n(t)]^2\rangle$ as a function of $\tau = 2\lambda^{(2)}t$.

A. Fokker-Planck Equations and Two-Photon Absorption Processes

Recently, a general technique was found for the treatment of quantum-dynamical problems in phase space.[34] We can use this technique to obtain the dynamical equation obeyed by the phase space equivalent of the density operator. One finds by straightforward, but long, calculations that the antinormally ordered equivalent[23] of the density operator obeys the following equation of motion:

$$\begin{aligned}\frac{\partial F_\rho^{(A)}}{\partial t} = &-\lambda^{(2)}\left[(\kappa_1-\kappa_2)\frac{\partial^2}{\partial z^2}(z^2F_\rho^{(A)})+\text{c.c.}\right]+2\lambda^{(2)}\kappa_2\frac{\partial^4 F_\rho^{(A)}}{\partial z^2\partial z^{*2}}+\lambda^{(2)}\left[2\kappa_1\frac{\partial}{\partial z}(z^2z^*F_\rho^{(A)})+\text{c.c.}\right]\\ &+8\lambda^{(2)}\kappa_2\frac{\partial^2}{\partial z\partial z^*}[(zz^*+1)F_\rho^{(A)}]-\kappa_2\lambda^{(2)}\left[\frac{\partial}{\partial z}\left\{(2z^2z^*+4z)F_\rho^{(A)}\right\}+\text{c.c.}\right]\\ &-\lambda^{(2)}\kappa_2\left[4\frac{\partial^3}{\partial z^*\partial z^2}(zF_\rho^{(A)})+\text{c.c.}\right].\end{aligned} \tag{4.47}$$

This equation contains the entire information about the statistics of the photon system in two-photon absorption processes. At zero temperature, Eq. (4.47) reduces to

$$\frac{\partial F_\rho^{(A)}}{\partial t} = -\lambda^{(2)}\left[\frac{\partial^2}{\partial z^2}(z^2F_\rho^{(A)})+\text{c.c.}\right]+2\lambda^{(2)}\left[\frac{\partial}{\partial z}(z^2z^*F_\rho^{(A)})+\text{c.c.}\right]. \tag{4.48}$$

This equation can be used to calculate the *normally ordered time-ordered correlation functions* in two-photon absorption processes provided one can solve it for the Green's function.[35] When $\lambda^{(2)}$ is small, Eq. (4.48) can be solved by perturbation. By transforming (4.48), one can also obtain the stochastically equivalent Langevin equation[36]

$$\dot{z} = \lambda^{(2)}[z-2z^2z^*]+i\sqrt{\lambda^{(2)}}zf(t), \tag{4.49}$$

where the stochastic force $f(t)$ is a δ-correlated Gaussian random process, characterized by

$$\langle f(t)f^*(t')\rangle = 0,\ \langle f(t)\rangle = \langle f^*(t)\rangle = 0, \tag{4.50}$$

$$\langle f(t)f(t')\rangle = \langle f^*(t')f^*(t)\rangle = 2\delta(t-t'). \tag{4.51}$$

All the higher-order linked moments are equal to zero. Thus, we conclude that the change in the statistical properties of the photon system in two-photon absorption processes may be described by the nonlinear Langevin equation (4.49).

So far we have not taken into account the field losses by any process which is not a direct interaction between the field mode and the medium. We may represent the loss mechanism with an ensemble of harmonic oscillators in thermal equilibrium at some temperature T_B. Then the losses can be taken into account by adding to the right-hand side of (4.47) the following terms:

$$\mathcal{L}_1 F_\rho^{(A)} = 2\pi h(\omega) g_0^2(\omega) \langle n(\omega)\rangle \frac{\partial^2 F_\rho^{(A)}}{\partial z \partial z^*} + \pi h(\omega) g_0^2(\omega) \left\{ \frac{\partial}{\partial g} (z F_\rho^{(A)}) + \text{c. c.} \right\} \quad (4.52)$$

where $\langle n(\omega)\rangle = 1/\{e^{\hbar\omega/kT_B} - 1\}$. (4.53)

Here $g_0(\omega)$ is the coupling constant between the loss mechanism and the field mode and $h(\omega)$ is the density of the harmonic oscillators in terms of which the loss mechanism is described. The details of (4.52) can be found in Ref. 30.

In this paper, we have considered only multiphoton absorption processes. However, it seems that our method can be extended, with minor modifications, to other multiphoton processes (cf. Refs. 6, 11, and 12).

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[27]See, for example, Ref. 24, Chap. IX.

[28]See, for example, Ref. 25, p. 33.

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