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Theory of Two-Photon Absorption

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Abstract A theory of two-photon absorption from polychromatic stationary radiation field is presented. It is found that the transition probability of two-photon absorption depends on the statistical properties of the light employed. Namely, the transition probability for a coherent light is somewhat larger than that for an incoherent one.

I. Introduction

It is very interesting to consider whether the statistical properties of the radiation field affect the two-photon rate or not. Numerous reports have been presented on this problem[1)-5)]. For instance, a theory of the two-photon absorption from a single mode of the radiation field was described by Lambropoulos *et al.* (1966). They showed that assuming the same intensity for incoherent and coherent fields, the transition probability for incoherent light is twice as high as that for coherent one[3)]. For polychromatic modes of the field, another theory was described by Carusotto *et al.* (1967); they suggested that the results from a single mode of the field have no direct applications since almost experiments have been performed by polychromatic lights, and they showed that the two-photon absorption probability depends on the statistical properties of the light employed[4)]. Guccione *et al.* pointed out that Carusotto *et al.* compared the transition probability for a light pulse generated by a laser with that for thermal origin. Guccione *et al.* developed a polychromatic field theory and drew the conclusion that making use of polychromatic light, there is no distinction between the transition probability for coherent light and that for incoherent one[5)].

As will be shown later on, the two-photon absorption probability is proportional to a certain correlation function, while the properties of the field is included in a statistical distribution function which is incorporated in the correlation function. We expect that

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the difference in nature (i. e., phase and photon distribution) between a coherent light and an incoherent one, will affect the two-photon absorption process.

In addition to the reexamination of the above problem, we are particularly interested in dealing theoretically with the two-photon absorption from two independent light sources, a coherent light source and incoherent one, illuminating the sample system simultaneously. A few experiments have been reported for this technique[6)-8)].

Calculations are performed by means of the usual quantum mechanical perturbation theory; and our procedures are similar to those described by Guccione *et al*[5)]. The state of the radiation field is described by the density operator, the basis for the state description being constructed with the coherent states.

II. General Theory

In the presence of the radiation field, the Hamiltonian of the system is described by

$$H = H_r + H_p + H_{int} = H_0 + H_{int},\ H_0 = H_r + H_p, \tag{1}$$

where H_r refers to the radiation field and H_p to the particles. H_0 is the Hamiltonian in the absence of interaction between the particle and the field. H_r is written as

$$H_r = \sum_k \omega_k a_k^+ a_k, \tag{2}$$

where a_k^+ and a_k are the creation and annihilation operators for the k-th mode in the field, respectively. ω_k is the frequency of a photon of mode k. In the present paper, we put $\hbar = c = 1$, for convenience. a_k^+ and a_k satisfy the following commutation relations, $[a_k, a_k^{+\prime}] = \delta_{kk'}$, $[a_k, a_k{}']_- = [a_k^+, a_k^{+\prime}]_- = 0$. H_p is written as

$$H_p = \sum_s \epsilon_s c_s^+ c_s, \tag{3}$$

where c_s^+ and c_s are, respectively, the creation and annihilation operators for the s-th particle state. They satisfy the following commutation relations, $[c_s, c_r^+]_+ = \delta_{sr}$, $[c_s, c_r]_+ = [c_s^+, c_r^+]_+ = 0$.. ϵ_s is the energy of the particle in the state $|s\rangle$. H_{int} is the interaction Hamiltonian between the particle and the field, which is described in the nonrelativistic representation as

$$H_{int} = -\frac{e}{m} A \cdot P + \frac{e^2}{2m} A^2. \tag{4}$$

The symbols in the equation have the usual meaning; A is the vector potential operator in the Coulomb guage, P the momentum of the electron, and m and e are the mass and charge of the electron, respectively. As has been discussed by various authors, the A^2 term does not add any new features to the present problem of photon statistics[3),5)]. Thus, we ignore the A^2 term and shall discuss only the $A \cdot P$ term.

Now the vector potential A can be expanded in terms of the plane waves

$$A = \left(\frac{2\pi}{V}\right)^{1/2} \sum_k \omega_k^{1/2}\, \boldsymbol{e}_k\, (a_k\, e^{ik\cdot r} + adjoint), \tag{5}$$

where V is the quantization volume, and $\boldsymbol{k}$ and e_k are the wave vector and polarization vector, respectively. Substitution of Eq. (5) into Eq. (4) gives

$$H_{int} = \sum_{srk} (\, v_{sr}^k\, a_k + v_{sr}^{*k}\, a_k^+) c_s^+\, c_r, \tag{6}$$

where

$$v_{sr}^k = -\left(\frac{2\pi e^2}{m^2 \omega_k}\right) \langle s \mid \boldsymbol{p} \cdot \boldsymbol{e}_k\, exp(i\boldsymbol{k}\cdot\boldsymbol{r}) \mid r \rangle. \tag{7}$$

The coefficients v_{sr}^k may be evaluated by introducing the truncation of $e^{ik\cdot r}$ up to the dipolar term, since the wavelength of the radiation must be long compared to the size of the atom.

The density operator ρ_i in the interaction picture before the interaction, namely at t=0, is related to the density operator ρ_f after the interaction as

$$\rho_f = U\,(t)\, \rho_i\, U^+(t), \tag{8}$$

where $U(t) = U(t,\, 0)$ is the time developement operator. In Eq. (8) are included any informations for the all transitions from the initial state. The operator $U(t)$ satisfies a differential equation

$$i\frac{\partial}{\partial t}\, U(t,\, 0) = H_I U(t,\, 0), \tag{9}$$

and integrating the equation with respect to time gives

$$U(t,\, 0) = 1 - i\int_0^t H_I(\tau) U(\tau)\, d\,\tau, \tag{10}$$

here $H_I(t)$ is defined by

$$H_I(t) = exp\,(iH_0 t) H_{int}\; exp(-iH_0 t). \tag{11}$$

This equation is combined with Eq. (7) to give

$$H_I(t) = \sum_{srk} \{ v_{sr}^k a_k exp(-i\,\omega_k t) + adjoint \}\;\; c_s^+\, c_r\, exp(i\,\omega_{sr}\, t), \tag{12}$$

where we have used the relations

$$\begin{gathered} exp(iH_r t) a_k\; exp(-i\,H_r t) = a_k exp\,(-i\,\omega_k t), \\ exp\;(iH_s t) c_s^+ c_r\; exp(-iH_s t) = c_s^+ c_r exp(i\,\omega_{sr} t), \\ \omega_{sr} = \omega_s - \omega_r. \end{gathered} \tag{13}$$

The solution of the time developement operator takes the form

$$U(t)=1+(-i)\int_0^t H_I(\tau)d\tau+(-i)^2\int_0^t d\tau\int_0^\tau d\tau' H_I(\tau)H_I(\tau')+\cdots\cdots. \tag{14}$$

The second term of Eq. (14) is concerned with the one-photon transition, the third term with the two-photon transition, and so on. As we are interested in the two-photon process, we consider only the third term and denote it by $U^{(2)}(t)$. Substitution of Eq. (6) into Eq. (14) yields

$$U^{(2)}(t)=(-i)^2\int_0^t d\tau\int_0^\tau d\tau' \sum_{s,r,k_1}\sum_{s',r',k_2} v_{sr}^{k_1} v_{s'r'}^{k_2} a_{k1}\, a_{k2} c_s^+ c_r c_{s'}^+ c_{r'} \tag{15}$$
$$\times\, exp(-i\omega_{k_1}\tau) exp\,(-i\omega_{k_2}\tau').$$

Before interaction the atom and field are uncoupled, and one can assume that

$$\rho_i=\rho_{pi}\,\rho_{ri}, \tag{16}$$

where $\rho_{pi}=|i\rangle\langle i|$ is the density operator of the atom in the initial state, and ρ_{ri} is that of the field. In order to describe the radiation field, we introduce the following coherent states $|\alpha_k\rangle$ as the basis,

$$a_k\,|\,\alpha_k\rangle=\alpha_k\,|\,\alpha_k\rangle, \tag{17}$$

where the range of the eigenvalue α_k is the entire complex plane and α_k is in general a complex number. Similarly the multi-mode coherent states are defined by the products of single mode coherent stats, i.e.[9],

$$|\{\alpha_k\}\,\rangle=\Pi_k\,|\,\alpha_k\rangle. \tag{18}$$

The density operator ρ_{ri} can be expanded with these eigenstates

$$\rho_{ri}=\int\cdots\int P(\{\alpha_k\})\,\Pi_k\,|\,\alpha_k\rangle\langle\alpha_k\,|\,d^2\alpha_k, \tag{19}$$

where $d^2\alpha\equiv d\,[Re\,\alpha]\,d\,[Im\,\alpha]$,
and the function $P(\{\alpha_k\})$ has been called "P representation", which may be thought of as a weight function that characterizes the radiation field. Thus, the density operator before interaction ρ_i is obtained. The transition probability for two-photon absorption is obtained by tracing of Eq. (8) over both the field states and the final state of the atom. Thus, we introduce the reduced density matrix, $\chi^{(2)}(t)$, which has already been performed a trace over the fields states[5]. Then, the transition probability from the initial state to the final state becomes

$$\langle f\,|\,\chi^{(2)}(t)\,|\,f\rangle=\sum_{k_1,k_2,k_3,k_4}\{v_{fj}^{k_1} v_{ji}^{k_2} v_{li}^{*k_3} v_{fl}^{*k_4}(\omega_{k_2}-\omega_{ji})^{-1}(\omega_{k_3}-\omega_{li})^{-1}\}$$
$$\times\,\{exp[-i(\omega_{k_1}+\omega_{k_2}-\omega_{k_3}-\omega_{k_4})t/2]\frac{sin\,(\omega_{k_1}+\omega_{k_2}-\omega_{fi})t/2}{(\omega_{k_1}+\omega_{k_2}-\omega_{fi})/2} \tag{20}$$

$$\times \frac{\sin\,(\omega_{k_3}+\omega_{k_4}-\omega_{fi})\;{}^{t}/_{2}{}'}{(\omega_{k_3}+\omega_{k_4}-\omega_{fi})/2}\Big\}\int\cdots\int P(\{\alpha_k\})\,\alpha_{k_1}\,\alpha_{k_2}\,\alpha^*_{k_3}\,\alpha^*_{k_4}\Pi_k d^2\alpha_k.$$

It should be noted that only the last integration factor in Eq. (20) depends on the statistical properties of the light employed. Therefore the transition probability for two-photon absorption is proportional to the integration factor in Eq. (20), which corresponds to the second order correlation function introduced by Glauber[9].

III. Transition Probability for Stationary Field

In this section we shall compare the transition probability for a coherent light with that for an incoherent one. We assume that the radiation field is stationary and the transition rate given by Eq. (20) increases linearly with time for short times. The transition probability is then given by

$$\langle f\mid\chi^{(2)}(t)\mid f\rangle=2\pi t\sum_{k_1,k_2,k_3,k_4} M_{k_1k_2k_3k_4}\delta(\omega_{k_1}+\omega_{k_2}-\omega_{fi})$$
$$\times\,\delta(\omega_{k_1}+\omega_{k_2}-\omega_{k_3}-\omega_{k_4})\int\cdots\int P(\{\alpha_k\})\alpha_{k_1}\,\alpha_{k_2}\alpha^*_{k_3}\alpha^*_{k_4}\,\Pi_k d^2\alpha_k, \qquad (21)$$

where we denote the terms in the first set of braces in Eq. (20) by the symbol $M_{k1\,k2\,k3\,k4}$, in which all atomic informations are included. With the condition $t\gg\omega_{k}^{-1}$, the terms in the second set of braces in Eq. (20) are replaced by the δ functions. As the first of the δ functions is not applicable to the infinitely sharp energy levels, we introduce a finite width in the state density of the final state $\rho\,(\epsilon_f)$ to avoid the difficulty, and define a new transition probability $w(t)$:

$$w\;(t)\equiv\int\langle f\mid x^{(2)}(t)\mid f\rangle\rho(\epsilon_f)d\,\epsilon_f$$
$$=2\pi t\sum_{k_1,k_2,k_3,k_4} M_{k_1k_2k_3k_4}\rho(\omega_{k_1}+\omega_{k_2}+\epsilon_i) \qquad (22)$$
$$\times\,\delta\,(\omega_{k_1}+\omega_{k_2}-\omega_{k_3}-\omega_{k_4})\int\cdots\int P(\{\alpha_k\})\alpha_{k_1}\,\alpha_{k_2}\,\alpha^*_{k3}\,\alpha^*_{k4}\prod_k d^2\alpha_k.$$

Now we recall that we can discriminate a coherent light from an incoherent one through the last integration factor in Eq. (22). We shall evaluate the transition probability both for a coherent light and incoherent one.

For a coherent light we assume that the weight function takes the following form

$$P(\{\alpha_k\})=\frac{1}{2\pi}\int_0^{2\pi}\prod_k p(\alpha_k)d\,\bar{\theta}, \qquad (23)$$

where $P(\alpha_k)=\delta^{(2)}(\mid\alpha_k\mid e^{i\bar{\theta}}-\alpha_k)$,

where $\delta^{(2)}(\alpha)=\delta\,(Re\,\alpha)\,\delta\,(Im\,\alpha)$.

The $\bar{\theta}$ integration in Eq. (23) means complete ignorance of the phase of the high frequency field (Glauber 1963). Substituting Eq. (23) into Eq. (22) and performing the integration, we get

$$
\begin{aligned}
w(t)= & 2\pi t \sum_{k_1} M_{k_1 k_1 k_1 k_1} \rho(2\omega_{k_1}+\epsilon_i)\langle n_{k_1}\rangle^2 \\
& +4\pi t \sum_{k_1,k_2}' M_{k_1 k_2 k_1 k_2} \rho(\omega_{k_1}+\omega_{k_2}+\epsilon_i)\langle n_{k_1}\rangle\langle n_{k_2}\rangle \\
& +2\pi t \sum_{k_1,k_2,k_3,k_4}' M_{k_1 k_2 k_3 k_4} \rho(\omega_{k_1}+\omega_{k_2}+\epsilon_i) \\
& \times \delta(\omega_{k_1}+\omega_{k_2}-\omega_{k_3}-\omega_{k_4})\langle n_{k_1}\rangle^{1/2}\langle n_{k_2}\rangle^{1/2}\langle n_{k_3}\rangle^{1/2}\langle n_{k_4}\rangle^{1/2},
\end{aligned}
\tag{24}
$$

where $\langle n_k\rangle \equiv T_r\{\rho a_k^+ a_k\} = |\alpha_k|^2$ is the average number of photons in the field. The symbol, $\sum_{k_1,k_2}'$, denotes excluding the case of $k_1=k_2$, while the symbol $\sum_{k_1,k_2,k_3,k_4}'$ means restricting to the cases where k_1, k_2, k_3, and k_4 differ from each other.

On the other hand, for an incoherent light, we assume that the weight function takes a Gaussian distribution. If we write $\alpha_k = |\alpha_k| e^{i\theta_k}$, the integration factor in Eq. (22) is given by

$$
\int\cdots\int P(\{\alpha_k\})\, |\alpha_{k_1}||\alpha_{k_2}||\alpha_{k_3}^*||\alpha_{k_4}^*|\, e^{i(\theta_{k_1}+\theta_{k_2}-\theta_{k_3}-\theta_{k_4})} \prod_k d^2\alpha_k,
\tag{25}
$$

where in order to make the integration nonvanishing, the phase factor of the exponential part of the integrand in Eq. (25) must be equal to zero. The following cases satisfy the above phase condition:

$$
\begin{aligned}
&(1)\quad \theta_{k_1}=\theta_{k_2}=\theta_{k_3}=\theta_{k_4} \text{ or } k_1=k_2=k_3=k_4, \\
&(2)\quad \theta_{k_1}=\theta_{k_3},\ \theta_{k_2}=\theta_{k_4} \text{ or } k_1=k_3,\ k_2=k_4, \\
&(3)\quad \theta_{k_1}=\theta_{k_4},\ \theta_{k_2}=\theta_{k_3} \text{ or } k_1=k_4,\ k_2=k_3, \\
&(4)\quad \theta_{k_1}+\theta_{k_2}=\theta_{k_3}+\theta_{k_4} \text{ and } k_1\neq k_2\neq k_3\neq k_4.
\end{aligned}
\tag{26}
$$

As assumed above the weight function $P(\{\alpha_k\})$ for an incoherent light takes a Gaussian distribution

$$
P(\{\alpha_k\}) = \prod_k \frac{1}{\pi\langle n_k\rangle} exp\,(-|\alpha_k|^2/\langle n_k\rangle).
\tag{27}
$$

Substituting Eq. (27) into Eq. (25) and invoking Eq. (26), we have

$$
\begin{aligned}
w(t)= & 4\pi t \sum_{k_1} M_{k_1 k_1 k_1 k_1} \rho(2\omega_{k_1}+\epsilon_i)\langle n_{k_1}\rangle^2 \\
& +4\pi t \sum_{k_1,k_2}' M_{k_1 k_2 k_1 k_2} \rho(\omega_{k_1}+\omega_{k_2}+\epsilon_i)\langle n_{k_1}\rangle\langle n_{k_2}\rangle \\
& +2\pi t \sum_{k_1,k_2,k_3,k_4}' M_{k_1 k_2 k_3 k_4} \rho(\omega_{k_1}+\omega_{k_2}+\epsilon_i)\left(\frac{\pi}{32}\right)^2 \\
& \times \delta(\omega_{k_1}+\omega_{k_2}-\omega_{k_3}-\omega_{k_4})\langle n_{k_1}\rangle^{1/2}\langle n_{k_2}\rangle^{1/2}\langle n_{k_3}\rangle^{1/2}\langle n_{k_4}\rangle^{1/2}.
\end{aligned}
\tag{28}
$$

In Eq. (24) and Eq. (28), the first terms reproduce the single mode contributions derived by Lambropoulos *et al.* It is clear that for the absorption from a single mode of the radiation field, the transition probability for an incoherent light is twice as high as that for a coherent one. The second and the third terms in Eq. (24) give the multi-mode contributions for a coherent light and those in Eq. (28) give the multi-mode contributions for an incoherent one. Guccione *et al.* compared the second term in Eq. (24) with that of Eq. (28) and concluded that there is no distinction between a coherent light and an incoherent light with respect to their transition probabilities for multi-mode of the field. However, if we assume the weight functions as given by Eq. (23) and Eq. (27), the third terms in Eq. (24) and Eq. (28) shoud emerge, which have not been found by Guccione et al. Namely, the transition probability of two-photon absorption depends on the statistical properties of the field.

Later, in section V, we shall show that the third term in Eq. (28) equals zero as the corresponding correlation function vanishes. Therefore we neglect the third term in Eq. (28). Next, considering the spectral width of the radiation field, we shall perform the summation in Eq. (24) and Eq. (28) ignoring the contributions from a single mode of the radiation field. As an example, assming a square spectral shape, we shall evaluate $w(t)$ for the two situations; (i) the spectral width of the radiation field is narrow compared with the width of the final state $\Delta\epsilon$, and (ii) the reverse case of (i).

(i) $\Delta\omega \gg \Delta\epsilon$

By fixing the mode k_2, the summation of the second term in Eq. (24) and that in Eq. (28) can be carried out convenientry; the number of mode k_1 such that the sum $\omega_{k_1}+\omega_{k_2}$ falls within the band of the final state equals $\frac{\Delta\epsilon}{\delta\omega}$, where $\delta\omega$ is the mode spacing. In addition, we assume that the spectrum is centered on the frequency $\omega_0=\frac{\omega_{fi}}{2}$. On the other hand, the summation of the third term in Eq. (24) is performed under the conditions that k_1+k_2 equals k_3+k_4 and that the sum $\omega_{k_1}+\omega_{k_2}$ falls within the band of the final state. Consequently we get

$$w(t)=2\pi t M_0(\Delta\omega)^{-1}\left(\frac{E}{\omega_0}\right)^2 N \text{ (coherent field)},$$

$$w(t)=4\pi t M_0(\Delta\omega)^{-1}\left(\frac{E}{\omega_0}\right)^2 \text{ (incoherent field)}. \tag{29}$$

where we assume that $\langle n_{k_1}\rangle=\langle n_{k_2}\rangle=\langle n_{k_3}\rangle=\langle n_{k_4}\rangle=\langle n\rangle$ is the number of photons per mode. N is the number modes in the radiation spectrum defined by $\Delta\omega/\delta\omega$, and E is the total energy of the field given by $N\langle n\rangle\omega_0$.

M_0 represents $M_{k_1k_2}$ or $M_{k_1k_2k_3k_4}$ as the case may be, evaluated under the frequency condition

$$\omega_{k_1}=\omega_{k_2}=\omega_{k_3}=\omega_{k_4}=\omega_0.$$

(ii) $\Delta\omega \ll \Delta\epsilon$

In this case, there is no restriction on k_1, k_2, k_3 and k_4, one of which is dummy in the summation in Eq. (24) because of the occurence of the δ function. Thus, summing over these k's from 1 to N, we get

$$w(t)=2\pi t M_0(\Delta\epsilon)^{-1}\left(\frac{E}{\omega_0}\right)^2 N \text{ (coherent field)},$$

$$w(t)=4\pi t M_0(\Delta\epsilon)^{-1}\left(\frac{E}{\omega_0}\right)^2 \text{ (incoherent field)}. \tag{30}$$

Therefore, the transition for a stationary coherent light is $N/2$ times as large as that for an incoherent one.

IV. Simultaneous Application of Coherent and Incoherent Light Sources

In this section, we shall consider the two-photon absorption induced by the simultaneous application of a coherent light and an incoherent one; the frequency of the coherent light is assumed to be too small to induce the two-photon absorption by itself, and the intensity of the incoherent light is assumed to be too weak to induce the absorption. Eq. (22) can be recast as follows

$$w(t)=2\pi t \sum_{k_1,k_2,k_3,k_4} M_{k_1k_2k_3k_4}\,\rho(\omega_{k_1}+\omega_{k_2}+\epsilon_i)\delta(\omega_{k_1}+\omega_{k_2}-\omega_{k_3}-\omega_{k_4})$$
$$\times\int\cdots\int\prod_k d^2\alpha_k P(\{\alpha_k\})\,|\alpha_{k_1}|\,|\alpha_{k_2}|\,|\alpha^*_{k_3}|\,|\alpha^*_{k_4}|\,e^{i(\theta_{k_1}+\theta_{k_2}-\theta_{k_3}-\theta_{k_4})}, \tag{31}$$

where, as a matter of course, either k_1 or k_2 refers to the coherent field, and either k_3 or k_4 refers to the coherent field. Assuming the weight function as

$$P(\{\alpha_k\})=\delta^{(2)}(|\alpha_{k_1}|e^{i\theta_{k_1}}-\alpha_{k_1})\delta^{(2)}(|\alpha_{k_3}|e^{i\theta_{k_3}}-\alpha_{k_3})$$
$$\times(\Pi\langle n_{k_2}\rangle)^{-1}e^{-|\alpha_{k_2}|^2/\langle n_{k_2}\rangle}(\Pi\langle n_{k_4}\rangle)^{-1}e^{-|\alpha_{k_4}|^2/\langle n_{k_4}\rangle}, \tag{32}$$

and substituting Eq. (32) into Eq. (31) we have

$$w(t)=4\pi t\sum_{k_1,k_2} M_{k_1k_2k_1k_2}\,\delta(\omega_{k_1}+\omega_{k_2}+\epsilon_i)\langle n_{k_1}\rangle\langle n_{k_2}\rangle. \tag{33}$$

It may be noted that Eq. (33) has the same form as the second term in Eq. (28) obtained for the application of an incoherent light. We can see that the transition probability for anyone of the two light sources.

V. Discussion

Certain correlation functions play the dominant role in the two-photon absorption process from the viewpoint of the coherent properties of light. According to Glauber. the n-th order correlation function is defined as[9]

$$g^{(n,n)}(k_1,\cdots k_n\,;\,k_{n+1}\cdots k_{2n})\equiv\langle\prod_{r=1}^{n}a^+_{k_r}\prod_{s=n+1}^{2n}a_{k_s}\rangle \tag{34}$$
$$\equiv T_r\,|\,\rho\prod_{r=1}^{n}a^+_{k_r}\prod_{s=n+1}^{2n}a_{k_s}\}\;.$$

In the above equation the properties of the field is incorporated in the density operator ρ, which is defined by $\sum_{n,m} | n\rangle\langle m | \rho_{nm}$ in the n representation for the pure coherent field, and by $\sum_{n} | n\rangle\langle n | \rho_{nn}$ for the incoherent one. The n-th order correlation function is decomposed as follows

$$g^{(n,n)}(k_1,\cdots k_n\,;\,k_{n+1}\cdots k_{2n})=\prod_{j=1}^{2n} g^{(1,1)}(k_j,\,k_j') \text{ (coherent field),} \tag{35}$$

$$g^{(n,n)}(k_1,\cdots k_n\,;\,k_{n+1}\cdots k_{2n})=\sum_{P}\prod_{j=1}^{n} g^{(1,1)}(k_j,\,k_j') \text{ (incoherent field),} \tag{36}$$

where the subscript P in Eq. (36) means that the summation should be carried out over $n!$ permutations. From the n-th order correlation function in Eqs. (35) and (36), we obtain the second order correlation functions

$$g^{(2,2)}(k_1,\,k_2\,;\,k_3,\,k_4)=\langle a^{+}_{k_1}\,a^{+}_{k_2}\,a_{k_3}a_{k_4}\rangle \text{ (coherent field),}$$

$$g^{(2,2)}(k_1\,k_2\,;\,k_3,\,k_4)=\langle a^{+}_{k_1}\,a_{k_3}\rangle\langle a^{+}_{k_2}a_{k_4}\rangle \tag{37}$$

$$+\langle a^{+}_{k_1}\,a_{k_4}\rangle\langle a^{+}_{k_2}\,a_{k_3}\rangle \text{ (incoherent field).}$$

For the incoherent field, where ρ is a diagonal matrix, we have the relation

$$\langle a^{+}_{k_i}\,a_{k_j}\rangle = |\,\alpha_{k_i}\,|^2\,\delta_{ij},$$

whereas, for the coherent field,

$$\langle a^{+}_{k_i}\,a_{k_j}\rangle = \alpha^{*}_{k_i}\,\alpha_{k_j}.$$

Therefore Eq. (37) becomes

$$g^{(2,2)}(k_1,\,k_2\,;\,k_3,\,k_4) = \alpha_{k_1}\,\alpha_{k_2}\,\alpha^{*}_{k_3}\,\alpha^{*}_{k_4} \text{ (coherent field),} \tag{38}$$

$$g^{(2,2)}(k_1,\,k_2\,;\,k_3,\,k_4) = 2\,|\,\alpha_{k_1}|^2\,|\alpha_{k_2}|^2 \text{ (incoherent field).} \tag{39}$$

The value given by Eq. (38) corresponds to the sum of the second and third terms in Eq. (24) and that given by Eq. (39) to the second term in Eq. (28). The third term in Eq. (28) equals zero for the corresponding correlation function vanishes. Therefore, comparing the transition probability for a coherent light with that for an incoherent light in Eq. (29) or Eq. (30), we find that the transition probability for a coherent light is $(N/2)$ times as large as that for an incoherent one, when the average number of the photon is the same, contrary to the results of Guccione *et al.* (1967). As mentioned in section III, the above finding is properly obtained as far as the weight function is assumed as Eq. (23) for a coherent light and as Eq. (27) for an incoherent light.

It is supposed that the effective number of field mode would not be so large in the laser source. Therefore we deduce that the transition probability for a laser may be somewhat larger than that for an incoherent light such as a Xenon lamp, of course assumig that the average number of photon in the field is the same. As essentially all experiments have been carried out by use of the nonstationary field (i. e., pulse) on the two-photon absorption, our results can not be compared with observations at present. However, in future, powerful stationary laser will be available, and we believe that our theory would offer some insights into the problem in the two-photon absorption concerned with the photon statistics.

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