Cooperative emission in the process of cascade and dipole-forbidden transitions

N. Enaki[1,2] and M. Macovei[1]
[1]Institute of Applied Physics, Academy of Sciences of Moldova, Academy Strasse 5, Kishinev MD-2028, Moldova
[2]Institute of Atomic Physics, IFIN, Magurele, Bucharest, Romania
(Received 10 February 1997)

In this paper the difference between two-photon dipole-forbidden and cascade transitions of an inverted system of atoms is studied. It is shown that the fluctuations of the electromagnetic field (EMF) density in the case of two-photon dipole-forbidden transitions are larger than the ones in the case of cascade emission. In the process of the two-photon dipole-forbidden cooperative emission the photons are created in pairs and there appears coherence between pairs of photons (biphotons). In this situation the distribution of energy between the photons in one pair may be random. For two-photon dipole-forbidden transitions the time-behavior of the square density fluctuations of EMF is similar to those of the density of EMF in the case of cascade emission. When the distance between two atoms excited relative to dipole-forbidden transitions is less than the radiation wavelength, then the kinetics of such atoms is analogous to the cascade emission of a single atom. [S1050-2947(97)09710-2]

PACS number(s): 42.50.Ct, 42.50.Lc, 32.80.−t

I. INTRODUCTION

The problem of quantum fluctuations and the generation of the nonclassical electromagnetic field in two-photon and multiphoton processes have recently been the subject of a number of theoretical and experimental studies [1–6]. From the physical point of view it is very interesting to study the behavior of the inverted system of radiators (nuclei, atoms) in the processes of the two-photon cooperative emission of light. For instance, the phenomenon of the new cooperative emission for a dipole-forbidden transition of inverted radiators can be observed in the process of the two-photon spontaneous emission [7,8]. It has been shown that in the process of spontaneous emission the atoms enter the regime of two-photon superradiance and the collective spontaneous rate of photon pairs (biphotons) increases proportionally to the square number of radiators. It has been demonstrated that in the process of spontaneous emission of hydrogen or helium-like atoms, the dipole-forbidden transitions $2^2S_{1/2}$-$1^1S_{1/2}$ and 2^2S_0-1^1S_0 generate pairs of correlated photons (biphotons). The correlation function between the pairs of photons becomes larger than that of individual photons from different pairs. A similar effect has been proposed in Ref. [9] in which the authors studied the exchange of the two-photon integral between two identical atoms separated by distance r_{12} (large compared to the atomic diameter, so the overlap is negligible). Such atoms undergo two-photon cooperative transitions between their two levels (of the same parity) via the nonresonant intermediate level (of the opposite parity).

It is known that Dicke one-photon superradiance has became the focus of extensive theoretical and experimental investigations [10–12]. The cooperative two-photon decay of the two-level system, inverted relative to the dipole-forbidden transition, is a more difficult effect, because of the electromagnetic field (EMF) exchange between the radiators taking place through biphotons. In this paper we study the difference between the two-photon dipole-forbidden cooperative emission of two atoms and the cascade emission of a single atom using the quantum statistical description of the radiation field. It is shown that in the case of cooperative dipole-forbidden transitions the photons in the radiation field form time-correlated pairs that for a large number of atoms leads to superbunching phenomena. The chain of equations for the atomic subsystem for the cases of both two-photon and two one-photon emission has been obtained. The exchange integral between the radiators through the EMF is analyzed in Secs. II and III. The kinetic characteristic for cooperative emission of four photons by two atoms situated at an arbitrary distance has been obtained. It has been shown that in the case when the distance between the two atoms, excited relative to the dipole-forbidden transition, is less than the emission wavelength, then the kinetics of such atoms is analogous to the cascade emission of two identical photons by one atom. Therefore the fluctuations of the light intensity in the case of two-photon dipole-forbidden transitions are larger than those of cascade emission.

The behavior of the inverted system of a large number of radiators has been analyzed too. It follows (from our results) that for the cooperative two-photon dipole-forbidden transition both the second- and the first-order correlation functions of the light intensity are proportional to the emission rate. In this situation we neglect the fluctuations of the number of excited atoms in the process of dipole-forbidden transition, which do not substantially change the fluctuation value of the EMF intensity. This approximation gives us the possibility to close the chain of equations for the atomic subsystem and enables us to study the time behavior of the quantum fluctuations of the EMF intensity.

In the case of cascade emission the first- and the second-order correlation functions for the light intensity of the cooperative emission are directly proportional to the emission rate and to the square emission rate operators of the atomic subsystem, respectively. In this situation neglecting the fluctuations of a number of excited atoms in the chain of atomic subsystem equations strongly affects the quantum fluctuation of a number of emission photons. Here, it should be mentioned that the relative fluctuations of the square intensity operator in the case of two-photon dipole-forbidden transitions have a time behavior similar to the relative fluctuations of the intensity operator for the cascade emission.

This paper is organized as follows. In Sec. II we propose a three-level Hamiltonian of inverted radiators for dipole-forbidden and cascade transitions. Using the method of eliminating operators of the EMF and the virtual state of electrons in the atoms we obtain master equations for the atomic subsystem. In Sec. III we express the correlation functions for the EMF intensity through the correlators of the atomic subsystem operators. This representation of the correlation functions gives us the possibility to study the time behavior of the quantum fluctuations of the EMF in the process of two-photon dipole-forbidden and cascade emissions. Such a time behavior of the inverted atomic subsystem is studied in Sec. IV.

II. MASTER EQUATION FOR THE ATOMIC SUBSYSTEM DENSITY MATRIX

We consider an ensemble of N three-level radiators in the exited state $|2\rangle$. Since the transitions between the $|2\rangle$ and $|3\rangle$ and $|1\rangle$ and $|3\rangle$ levels are allowed ($d_{32}, d_{31} \neq 0$), while the transition is forbidden between $|2\rangle$ and $|1\rangle$ ($d_{21}=0$), the Hamiltonian of such a system takes the form

$$H=\sum_k \hbar\omega_k a_k^\dagger a_k+\sum_{\alpha=1}^{3}\sum_{j=1}^{N}\hbar\omega_\alpha U_{j\alpha}^{\alpha} +i\sum_{\beta=1}^{2}\sum_k\sum_{j=1}^{N}(\vec{d}_{3\beta},\vec{g}_k)(a_k^\dagger e^{-i\vec{k}\cdot\vec{r}_j}-\text{H.c.})(U_{j3}^{\beta}+U_{j\beta}^{3}). \tag{1}$$

Here $\hbar\omega_\alpha(\alpha=1,2,3)$ is the energy of level α; $d_{3\beta}$ is the dipole moment of the transition between the states $|3\rangle$ and $|\beta\rangle(\beta=1,2)$ $a_k^\dagger$ and a_k are the creation and annihilation operators for the photons with the momentum $\hbar\vec{k}$, energy $\hbar\omega_k$, and polarization λ, $\vec{g}_k=(2\pi\hbar\omega_k/V)^{1/2}\vec{e}_\lambda$, $\vec{e}_\lambda$ is the photon polarization vector $\lambda=1,2$ and V is the EMF quantization volume, $U_{j\beta}^{3}$ is the corresponding operator of the transition between states $|3\rangle$ and $|\beta\rangle$ of the jth atom. The operators of the atomic subsystem and the EMF operators satisfy the commutation relations

$$[U_{j\beta}^{\alpha},U_{l\alpha'}^{\beta'}]=\delta_{j,l}[\delta_{\beta\beta'}U_{j\alpha'}^{\alpha}-\delta_{\alpha\alpha'}U_{j\beta}^{\beta'}],$$

$$[a_k,a_{k'}^\dagger]=\delta_{kk'},\quad [a_k^\dagger,a_{k'}^\dagger]=[a_k,a_{k'}]=0.$$

Here we shall discuss two types of three level systems: (a) level $|3\rangle$ is situated higher than the $|2\rangle$ and $|1\rangle$ levels [see Fig. 1(a)] and (b) level $|3\rangle$ is situated between the $|2\rangle$ and $|1\rangle$ levels [see Fig. 1(b)].

Let us now analyze the first case (a) when the atomic subsystem is inverted relative to the dipole-forbidden transition $|2\rangle$ and $|1\rangle$. As the unpopulated level $|3\rangle$ is situated higher than the populated level $|2\rangle$ the real transition between levels $|2\rangle$ and $|3\rangle$ do not take place due to the violation of the conservation energy law ($E_2-E_3-\hbar\omega_k \neq 0$, E_2 and E_3 are the energy of the levels $|2\rangle$ and $|3\rangle$, respectively, $\hbar\omega_k$ is the emitted photon energy). In this situation only a two-photon transition between the excited state $|2\rangle$ and ground state $|1\rangle$ is possible and takes place via the virtual state $|3\rangle$. In such dipole-forbidden transitions the operators that describe the transitions between $|2\rangle$ and $|1\rangle$ levels belong to su(2) algebra [13].

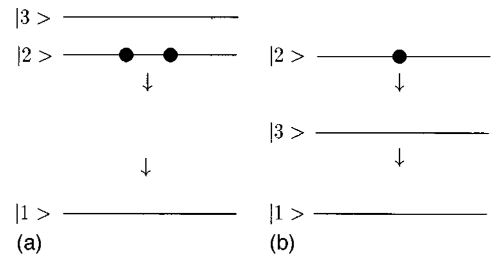

FIG. 1. (a) Initial state of the Λ-type three-level atomic system ($t=0$) in the case of two-photon dipole-forbidden transitions. (b) Initial state of cascade three-level atomic system ($t=0$) in the case of two one-photon emission.

Let us consider an operator $O(t)$, which describes the transition between levels $|2\rangle$ and $|1\rangle$. The Heisenberg equation for the operator $O(t)$ is given by the relation

$$\frac{d}{dt}O(t)=\frac{i}{\hbar}[H(t),O(t)]. \tag{2}$$

Using the Hamiltonian (1) one can obtain the following Heisenberg equation for the mean value of the operator $O(t)$:

$$\frac{d}{dt}\langle O(t)\rangle=\frac{i}{\hbar}\sum_{\alpha=1}^{3}\sum_{j=1}^{N}\hbar\omega_\alpha\langle[U_{j\alpha}^{\alpha}(t),O(t)]\rangle -\sum_k\sum_{j=1}^{N}\sum_{\beta=1}^{2}\frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)}{\hbar} \times\langle[U_{j\beta}^{3}(t)+U_{j3}^{\beta}(t),O(t)] \times(a_k^{+}(t)e^{-i\vec{k}\cdot\vec{r}_j}-\text{H.c.})\rangle, \tag{3}$$

where $\langle\,\rangle$ is averaging over the initial state of the ‘‘atom plus field’’ system $\Psi=|V\rangle|A\rangle|A\rangle$ is the wave function of the atomic subsystem for $t=0$, $|V\rangle$ is the wave function of the EMF vacuum). Equation (3) contains the EMF operators $a_k(t)$ and $a_k^\dagger(t)$. After integrating the Heisenberg equation for these operators we obtain the formal solution for $a_k(t)$ and $a_k^\dagger(t)$

$$a_k(t)=a_k^{v}(t)+a_k^{s}(t),\quad a_k^\dagger(t)=[a_k(t)]^\dagger, \tag{4}$$

where $a_k^{v}(t)=a_k^{v}(0)e^{-i\omega_k t}$ and

$$a_k^{s}(t)=\sum_{l=1}^{N}\sum_{\rho=1}^{2}\frac{(\vec{d}_{3\rho}\cdot\vec{g}_k)}{\hbar}e^{-i(\vec{k}\cdot\vec{r}_l)}\int_0^t d\tau \times e^{-i\omega_k\tau}[U_{l\rho}^{3}(t-\tau)+U_{l3}^{\rho}(t-\tau)]$$

are the vacuum and source parts of the operator $a_k(t)$. If we substitute the expressions (4) for $a_k(t)$ and $a_k^\dagger(t)$ into Eq. (3) we obtain the following equation for the mean value of the operator $O(t)$:

$$\frac{d}{dt}\langle O(t)\rangle = i\sum_{\alpha=1}^{3}\sum_{j=1}^{N}\omega_\alpha\langle[U_{j\alpha}^{\alpha}(t),O(t)]\rangle + \sum_k\sum_{j,l=1}^{N}\sum_{\rho,\beta=1}^{2}\frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)}{\hbar^2}$$
$$\times\Bigg(e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\{\langle[U_{j\beta}^{3}(t),O(t)]U_{l3}^{\rho}(t-\tau)\rangle+\langle[U_{j\beta}^{3}(t),O(t)]U_{l\rho}^{3}(t-\tau)\rangle+\langle[U_{j3}^{\beta}(t),O(t)]U_{l\rho}^{3}(t-\tau)\rangle$$
$$+\langle[U_{j3}^{\beta}(t),O(t)]U_{l3}^{\rho}(t-\tau)\rangle\}+\text{H.c.t.}[O^+(t)\rightarrow O(t)]\Bigg). \tag{5}$$

Here the H.c.t.$[O^+(t)\rightarrow O(t)]$ coincide with H.c. terms for the hermitian operators $[O(t)=O^+(t)]$, and for the nonHermitian operator $O(t)$ the notation H.c.t.$[O^+(t)\rightarrow O(t)]$ is equivalent to H.c. terms in which the $O^+(t)$ operator is replaced by $O(t)$.

It is not difficult to represent the operators $U_{j\alpha}^{\beta}$ through Bose operators of the atom $|\alpha\rangle$ and $|\beta\rangle$ states

$$U_{j\alpha}^{\beta}=C_{j\beta}^{\dagger}C_{j\alpha},\quad [C_{j\alpha},C_{l\beta}^{\dagger}]=\delta_{\alpha\beta}\delta_{jl},\quad [C_{j\alpha},C_{j\beta}]=[C_{j\alpha}^{\dagger},C_{j_\beta}^{\dagger}]=0.$$

In terms of Bose operators Eq. (5) takes the form

$$\frac{d}{dt}\langle O(t)\rangle = i\sum_{\alpha=1}^{3}\sum_{j=1}^{N}\omega_\alpha\langle[U_{j\alpha}^{\alpha},O(t)]\rangle+\{I_1+I_2+I_3+I_4+\text{H.c.t.}[O^+(t)\rightarrow O(t)]\}, \tag{6}$$

where

$$I_1=\sum_k\sum_{j,l=1}^{N}\sum_{\rho,\beta=1}^{2}\frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)}{\hbar^2}e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\langle C_{j3}^{\dagger}(t)[C_{j\beta}(t),O(t)]C_{l\rho}^{\dagger}(t-\tau)C_{l3}(t-\tau)\rangle,$$

$$I_2=\sum_k\sum_{j,l=1}^{N}\sum_{\rho,\beta=1}^{2}\frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)}{\hbar^2}e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\langle C_{j3}^{\dagger}(t)[C_{j\beta}(t),O(t)]C_{l3}^{\dagger}(t-\tau)C_{l\rho}(t-\tau)\rangle,$$

$$I_3=\sum_k\sum_{j,l=1}^{N}\sum_{\rho,\beta=1}^{2}\frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)}{\hbar^2}e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\langle C_{j3}(t)[C_{j\beta}^{\dagger}(t),O(t)]C_{l\rho}^{\dagger}(t-\tau)C_{l3}(t-\tau)\rangle,$$

$$I_4=\sum_k\sum_{j,l=1}^{N}\sum_{\rho,\beta=1}^{2}\frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)}{\hbar^2}e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\langle C_{j3}(t)[C_{j\beta}^{\dagger}(t),O(t)]C_{l3}^{\dagger}(t-\tau)C_{l\rho}(t-\tau)\rangle. \tag{7}$$

Account was taken that the $O(t)$ operator is a combination of Bose operators of $|2\rangle$ and $|1\rangle$ states.

As level $|3\rangle$ is situated higher than the excited state $|2\rangle$ the population of this level is equal to zero in the process of two-photon spontaneous emission.

Using the Hamiltonian (1) and the Heisenberg equation for $C_{j3}^{\dagger}(t)$ and $C_{j3}(t)$ operators it is very easy to represent these operators through $C_\alpha(t)$, $C_\alpha^{\dagger}(t)$ $(\alpha=1,2)$, and their free part

$$C_{j3}(t)=C_{j3}^{f}(t)+C_{j3}^{s}(t),$$

$$C_{j3}^{\dagger}(t)=[C_{j3}(t)]^{+}.$$

Here C_{j3}^{f} and C_{j3}^{s} are the free and source parts of operator C_{j3}, respectively:

$$C_{j3}^{f}(t)=C_{j3}(0)e^{-i\omega_3 t},\quad C_{j3}^{f}|3\rangle=0,$$

$$C_{j3}^{s}(t)=\sum_k\sum_{\gamma=1}^{2}\frac{(\vec{d}_{3\gamma}\cdot\vec{g}_k)}{\hbar}\int_0^t d\tau e^{-i\omega_3\tau}$$
$$\times[a_k^{\dagger}(t-\tau)e^{-i\vec{k}\cdot(\vec{r}_j)}-\text{H.c.}]C_{j\gamma}(t-\tau). \tag{8}$$

In this situation is necessary to exclude Bose operators C_{j3} and $C_{j3}^{\dagger}$ from Eq. (7). For this purpose it is necessary to use the following lemma:

Lemma. If Bose operators C_{j3} and $C_{j3}^{\dagger}$ lie between the two operators of the atomic subsystem $A(t_1)$ and $B(t_2)$ [$A(t_1)$,$B(t_2)$ do not contain the operators C_{j3} and $C_{j3}^{\dagger}$] belonging to other times, the elimination of the free part of these operators yields the following expression for the correlator:

$$\langle B(t_2)C_{j3}(t)A(t_1)\rangle=\langle B(t_2)C_{j3}^{s}(t)A(t_1)\rangle-e^{-i\omega_3(t-t_1)}\langle B(t_2)[C_{j3}^{s}(t_1),A(t_1)]\rangle,$$
$$\langle B(t_2)C_{j3}^{+}(t)A(t_1)\rangle=\langle B(t_2)C_{j3}^{\dagger s}(t)A(t_1)\rangle+e^{i\omega_3(t-t_2)}\langle[C_{j3}^{+s}(t_2),B(t_2)]A(t_1)\rangle. \tag{9}$$

The proof of this lemma for $C_{j3}(t)$ and $C_{j3}^{\dagger}(t)$ operators is similar to the demonstration proposed in [7] for the EMF operators. The commutators in Eq. (9) play the highest role in the two-photon spontaneous emission and only such commutators bring the main contribution to the two-photon process.

Then, after removing the free part of C_{j3} and $C_{j3}^{\dagger}$ operators using the previous lemma (see Appendix) we can obtain the following expressions for I_1,I_2,I_3,I_4 in the Born-Markoff approximation:

$$I_1\approx\sum_{k_1k_2}\sum_{j,l=1}^{N}\frac{(\vec{d}_{32}\cdot\vec{g}_{k_1})^2(\vec{d}_{31}\cdot\vec{g}_{k_2})^2}{\hbar^4(\omega_{k_1}+\omega_{32})^2}e^{i(\vec{k}_1+\vec{k}_2)\cdot(\vec{r}_j-\vec{r}_l)}\zeta(\omega_{k_2}+\omega_{k_1}-\omega_{21})\langle C_{j2}^{\dagger}(t)[C_{j1}(t),O(t)]C_{l1}^{\dagger}(t)C_{l2}(t)\rangle,$$

$$I_2\approx\sum_{k_1k_2}\sum_{j,l=1}^{N}\frac{(\vec{d}_{31}\cdot\vec{g}_{k_1})(\vec{d}_{32}\cdot\vec{g}_{k_1})(\vec{d}_{31}\cdot\vec{g}_{k_2})(\vec{d}_{32}\cdot\vec{g}_{k_2})}{\hbar^4(\omega_{k_1}+\omega_{32})(\omega_{k_1}-\omega_{31})}e^{i(\vec{k}_1+\vec{k}_2)\cdot(\vec{r}_j-\vec{r}_l)}\zeta(\omega_{k_2}+\omega_{k_1}-\omega_{21})$$
$$\times\langle C_{j2}^{\dagger}(t)[C_{j1}(t),O(t)]C_{l1}^{\dagger}(t)C_{l2}(t)\rangle,$$

$$I_3\approx\sum_{k_1k_2}\sum_{j,l=1}^{N}\frac{(\vec{d}_{31}\cdot\vec{g}_{k_1})(\vec{d}_{32}\cdot\vec{g}_{k_1})(\vec{d}_{31}\cdot\vec{g}_{k_2})(\vec{d}_{32}\cdot\vec{g}_{k_2})}{\hbar^4(\omega_{k_1}+\omega_{32})(\omega_{k_1}-\omega_{31})}e^{i(\vec{k}_1+\vec{k}_2)\cdot(\vec{r}_j-\vec{r}_l)}\zeta(\omega_{k_2}+\omega_{k_1}-\omega_{21})\langle[C_{j2}^{\dagger}(t),O(t)]C_{j1}(t)C_{l1}^{\dagger}(t)C_{l2}(t)\rangle,$$

$$I_4\approx\sum_{k_1k_2}\sum_{j,l=1}^{N}\frac{(\vec{d}_{31}\cdot\vec{g}_{k_1})^2(\vec{d}_{32}\cdot\vec{g}_{k_2})^2}{\hbar^4(\omega_{k_1}-\omega_{31})^2}e^{i(\vec{k}_1+\vec{k}_2)\cdot(\vec{r}_j-\vec{r}_l)}\zeta(\omega_{k_2}+\omega_{k_1}-\omega_{21})\langle[C_{j2}^{\dagger}(t),O(t)]C_{j1}(t)C_{l1}^{\dagger}(t)C_{l2}(t)\rangle. \tag{10}$$

Here $\zeta(\omega_{k_1}+\omega_{k_2}-\omega_{21})=\pi\delta(\omega_{k_1}+\omega_{k_2}-\omega_{21})+\mathrm{P}[i/(\omega_{k_1}+\omega_{k_2}-\omega_{21})]$, P indicates Cauchy principal value [14], ω_{31}, ω_{32}, ω_{21} are the transition frequences between states $|3\rangle\rightarrow|1\rangle$, $|3\rangle\rightarrow|2\rangle$, $|2\rangle\rightarrow|1\rangle$, respectively.

At last taking into account the fact that $U_{l2}^{1}=C_{l1}^{\dagger}C_{l2}$, $U_{j1}^{2}=C_{j2}^{\dagger}C_{j1}$, the final expression for the mean value of $O(t)$ operator is

$$\frac{d}{dt}\langle O(t)\rangle=\frac{i}{\hbar}\langle[H^{\mathrm{eff}}(t),O(t)]\rangle+\sum_{j,l=1}^{N}\{F(j,l)\langle[U_{j1}^{2}(t),O(t)]U_{l2}^{1}(t)\rangle+\text{H.c.t.}[O^{+}(t)\rightarrow O(t)]\}, \tag{11}$$

where

$$H^{\mathrm{eff}}(t)=H_0(t)+H_i(t),\quad H_0(t)=\sum_{j=1}^{N}\sum_{\alpha=1}^{2}\hbar\omega_\alpha U_{j\alpha}^{\alpha}(t),$$

$$H_i(t)=\frac{d_{31}^2d_{32}^2}{4\pi^2\hbar c^6}\sum_{j,l=1}^{N}\int_0^\infty d\omega_{k_1}\omega_{k_1}^3\int_0^\infty d\omega_{k_2}\omega_{k_2}^3\mathrm{P}\frac{1}{\omega_{k_1}+\omega_{k_2}-\omega_{21}}\left(\frac{1}{\omega_{31}-\omega_{k_1}}+\frac{1}{\omega_{32}+\omega_{k_1}}\right)^2\chi_{jl}(\omega_{k_1})\chi_{jl}(\omega_{k_2})U_{j1}^{2}(t)U_{l2}^{1}(t),$$

$$F(j,l)=\frac{d_{31}^2d_{32}^2}{4\pi^2\hbar^2c^6}\int_0^{\omega_{21}}\omega_k^3(\omega_{21}-\omega_k)^3d\omega_k\chi_{jl}(\omega_k)\chi_{jl}(\omega_{21}-\omega_k)\left(\frac{1}{\omega_{31}-\omega_k}+\frac{1}{\omega_{32}+\omega_k}\right)^2$$

and

$$\chi_{jl}(X)=(1-\cos^2\theta)\frac{\sin(Xr_{jl}/c)}{Xr_{jl}/c}+(1-3\cos^2\theta)\left(\frac{\cos(Xr_{jl}/c)}{(Xr_{jl}/c)^2}-\frac{\sin(Xr_{jl}/c)}{(Xr_{jl}/c)^3}\right).$$

Here θ is the angle between the direction of the dipole moment $\vec{d}_{3\beta}$ and $\vec{r}_{jl}$, $H_i(t)$ describes the two-photon cooperative Lamb shift of two level states of atoms in the process of the spontaneous decay of the excited state $|2\rangle$, $F(j,l)$ is the spontaneous cooperative emission rate for j and l atoms of the system in the process of two-photon dipole-forbidden transitions, $\vec{r}_{jl}=\vec{r}_j-\vec{r}_l$, $r_{jl}=|\vec{r}_{jl}|$ is the distance between the j and l atoms.

Now we can easily obtain the equation for the density matrix $\rho(t)$. As

$$\mathrm{Tr}\left(\frac{d}{dt}O(t)\rho(0)\right)=\mathrm{Tr}\left(\frac{d}{dt}\rho(t)O(0)\right), \tag{12}$$

from Eq. (11) we get

$$\frac{d}{dt}\rho(t)=\frac{i}{\hbar}[\rho(t),H^{\mathrm{eff}}]+\sum_{j,l=1}^{N}\{F(j,l)[U_{l2}^{1}\rho(t),U_{j1}^{2}]+\mathrm{H.c.}\}. \tag{12a}$$

From Eqs. (11) and (12a) follows that cooperation takes place between the photon pairs (biphotons). The energy of the biphotons is fixed, $\hbar\omega_{21}=\hbar\omega_{k1}+\hbar\omega_{k2}$. The common phases of such photon pairs (biphotons) are in phase in spite of the fact that the photons in the pair may have different energies (for example, $\hbar\omega_{k1}$ may be $\geqslant$ than $\hbar\omega_{k2}$). In other words the two-photon dipole-forbidden cooperative decay has a broad band emission spectrum of biphotons $\hbar\omega_k\in(0,\omega_{21})$ and the spectral generation rate is determined primarily by the multiple $\omega_k^3(\omega_{21}-\omega_k)^3$. Such behavior of the cooperative radiation field is not specific for the two one-photon cascade emision.

Now let us consider the second case (b) when the $|3\rangle$ level is situated between $|2\rangle$ and $|1\rangle$ levels [see Fig. 1(b)]. If the transition frequences and the dipole moments of the transitions between the states $|2\rangle\rightarrow|3\rangle$ and $|3\rangle\rightarrow|1\rangle$ are respectively equal, $\omega_{23}=\omega_{31}=\omega_0$, $d_{23}=d_{31}=d_0$, the Hamiltonian of such an inverted system takes a simpler form:

$$H(t)=\sum_{j=1}^{N}\hbar\omega_0 D_{zj}(t)+\sum_k\hbar\omega_k a_k^{\dagger}a_k+i\sum_{j=i}^{N}\sum_k\frac{(\vec{d}_0\cdot\vec{g}_k)}{2^{1/2}}[D_j^{+}(t)a_k(t)e^{i\vec{k}\cdot\vec{r}_j}-\mathrm{H.c.}]. \tag{13}$$

Here the operators $D_j^{+}(t)$, $D_j^{-}(t)$ are the transition operators between $|2\rangle$ and $|1\rangle$ states via the real $|3\rangle$ state and they differ from the $U_{1j}^{2}(t)$, $U_{2j}^{1}(t)$ operators defined for the dipole-forbidden case (a):

$$D_{zj}(t)=U_{2j}^{2}(t)-U_{1j}^{1}(t),$$

$$D_j^{+}(t)=2^{1/2}[U_{3j}^{2}(t)+U_{1j}^{3}(t)],$$

$$D_j^{-}(t)=2^{1/2}[U_{2j}^{3}(t)+U_{3j}^{1}(t)],$$

and satisfy the following commutation relations

$$[D_j^{+}(t),D_k^{-}(t)]=2D_z(t)\delta_{jk},$$

$$[D_{zj}(t),D_k^{\pm}(t)]=\pm D^{\pm}(t)\delta_{jk}.$$

Using Heisenberg representation for the $Q(t)$ operator belonging to the atomic subsystem and removing the $a(t)$ and $a^{\dagger}(t)$ Bose operators of the EMF we can obtain the following equation for $\langle Q(t)\rangle$:

$$\frac{d}{dt}\langle Q(t)\rangle=i\omega_0\sum_{j=1}^{N}\langle[D_{zj}(t),Q(t)]\rangle+\frac{d_0^2\omega_0^3}{2\hbar c^3}\sum_{j,l=1}^{N}\chi_{jl}(\omega_0)\{\langle[D_j^{+}(t),Q(t)]D_l^{-}(t)\rangle+\mathrm{H.c.t.}[Q^{+}(t)\rightarrow Q(t)]\}$$
$$-i\frac{d_0^2}{2\pi\hbar c^3}\sum_{j,l=1}^{N}\mathrm{P}\int_0^{\infty}\frac{\omega^3 d\omega}{\omega-\omega_0}\chi_{jl}(\omega)\langle[D_j^{+}(t)D_l^{-}(t),Q(t)]\rangle. \tag{14}$$

Following the transformations similar with those in Eq. (12a), we obtain from Eq. (14) the next master equation for the atomic subsystem:

$$\frac{d}{dt}\rho(t)=\frac{i}{\hbar}[\rho(t),H^{\mathrm{eff}}]+\frac{3}{8\tau_A}\sum_{j,l=1}^{N}\chi_{jl}(\omega_0)\{[D_j^{-}\rho(t),D_l^{+}]+\mathrm{H.c.}\}, \tag{15}$$

where $1/\tau_A=4\omega_0^3d_0^2/3\hbar c^3$ and

$$H^{\mathrm{eff}}=\hbar\omega_0\sum_{j=1}^{N}D_{zj}(t)-\frac{d_0^2}{2\pi c^3}\sum_{j,l=1}\mathrm{P}\int_0^{\infty}\frac{\omega^3 d\omega}{\omega-\omega_0}\chi_{jl}(\omega)D_j^{+}(t)D_l^{-}(t).$$

From master equations (12a) and (15) follows that the dissipation rate of the excited energy in the case of the two-photon spontaneous emission is proportional to the product of the two exchange integrals $\chi_{jl}(x)\chi_{jl}(\omega_{21}-x)$ but in the case of the two one-photon cascade emission the dissipation rate is proportional only to $\chi_{jl}(\omega)$.

In the next section we study the photon correlation functions in these two cases.

III. PHOTON CORRELATION

The general photoelectron counting correlation functions can be represented by the positive $[\vec{E}^-(\vec{r},t)]$ and negative $[E^+(\vec{r},t)]$ frequency parts of the $\vec{E}(\vec{r},t)$ operator [14–18].

$$G_1(\vec{r},t)=\langle[\vec{E}^-(\vec{r},t),\vec{E}^+(\vec{r},t)]\rangle, \tag{16a}$$

$$G_2(\vec{r},t)=\langle:[\vec{E}^-(\vec{r},t),\vec{E}^+(\vec{r},t)][\vec{E}^-(\vec{r},t),\vec{E}^+(\vec{r},t)]:\rangle, \tag{16b}$$

$$G_4(\vec{r},t)=\langle:[\vec{E}^-(\vec{r},t),\vec{E}^+(\vec{r},t)][\vec{E}^-(\vec{r},t),\vec{E}^+(\vec{r},t)][\vec{E}^-(\vec{r},t),\vec{E}^+(\vec{r},t)][\vec{E}^-(\vec{r},t),\vec{E}^+(\vec{r},t)]:\rangle. \tag{16c}$$

Here $G_2(\vec{r},t)$ and $G_4(\vec{r},t)$ are respectively the intensity and the square intensity correlation functions for the same time t,

$$\vec{E}^-(\vec{r},t)=\sum_k \vec{g}_k a_k^\dagger(t)e^{-i\cdot\vec{k}\vec{r}},\quad \overrightarrow{E}^+(\vec{r},t)=[\vec{E}^-(\vec{r},t)]^\dagger,$$

$:f(\vec{r},t):$ indicates normal ordering. The functions

$$\begin{aligned}\Lambda_2(\vec{r},t)&=G_2(\vec{r},t)-G_1^2(\vec{r},t),\\ \Lambda_4(\vec{r},t)&=G_4(\vec{r},t)-G_2^2(\vec{r},t)\end{aligned} \tag{16$'$}$$

take into account the EMF density and the square EMF density fluctuations.

Let us consider the first case (a) when the unpopulated level $|3\rangle$ is situated higher than the levels $|2\rangle$ and $|1\rangle$. After partial elimination of the photon operators we obtain the following expressions for G_1 and G_2 correlation functions:

$$\begin{aligned}G_1(\vec{r},t)=&\sum_{k_1k_2}(\vec{g}_{k_1}\vec{g}_{k_2})\sum_{j,l=1}^{N}\sum_{\alpha_1,\alpha_2}^{2}\frac{(\vec{d}_{3\alpha_1}\cdot\vec{g}_{k_1})(\vec{d}_{3\alpha_2}\cdot\vec{g}_{k_2})}{\hbar^2}e^{-i\vec{k}_2\cdot\vec{r}_l+i\vec{k}_1\cdot\vec{r}_j}\int_0^t d\tau_1 e^{i\omega_{k_1}\tau_1}\int_0^t d\tau_2 e^{-i\omega_{k_2}\tau_2}\\ &\times\langle[U_{j\alpha_1}^3(t-\tau_1)+U_{j3}^{\alpha_1}(t-\tau_1)][U_{3l}^{\alpha_2}(t-\tau_2)+U_{j\alpha_2}^3(t-\tau_2)]\rangle e^{-i(\vec{k}_1-\vec{k}_2)\cdot\vec{r}},\end{aligned} \tag{17}$$

$$\begin{aligned}G_2(\vec{r},t)=&\sum_{k_1\cdots k_4}(\vec{g}_{k_1}\cdot\vec{g}_{k_2})(\vec{g}_{k_3}\cdot\vec{g}_{k_4})\sum_{j,l=1}^{N}\sum_{\alpha_1,\alpha_2=1}^{2}\frac{(\vec{d}_{3\alpha_1}\cdot\vec{g}_{k_1})(\vec{d}_{3\alpha_2}\cdot\vec{g}_{k_2})}{\hbar^2}e^{i(\vec{k}_1\cdot\vec{r}_j-\vec{k}_2\cdot\vec{r}_l)}\int_0^t d\tau_1 e^{i\omega_{k_1}\tau_1}\int_0^t d\tau_2 e^{-i\omega_{k_2}\tau_2}\\ &\times\langle[U_{j\alpha_1}^3(t-\tau_1)+U_{j3}^{\alpha_1}(t-\tau_1)]a_{k_3}^\dagger(t)a_{k_4}(t)[U_{3l}^{\alpha_2}(t-\tau_2)+U_{l\alpha_2}^3(t-\tau_2)]\rangle e^{-i(\vec{k}_1+\vec{k}_3-\vec{k}_2-\vec{k}_4)\cdot\vec{r}}.\end{aligned} \tag{18}$$

We observe that it is difficult to exclude the EMF operators $a_{k_3}^\dagger(t)$ and $a_{k_4}(t)$ from Eq. (18) because the atomic subsystem operators $[U_{j\alpha_1}^3(t-\tau_1)+U_{j3}^{\alpha_1}(t-\tau_1)]$ and $[U_{l\alpha_2}^3(t-\tau_2)+U_{3l}^{\alpha_2}(t-\tau_2)]$ belong to other time moments. In this situation one can exclude the boson operators from these expressions using the similar lemma for the boson operators of EMF [7].

$$G_1(\vec{r},t)=\sum_{k_1k_2k_3}(\vec{g}_{k_1}\cdot\vec{g}_{k_2})\frac{(\vec{d}_{31}\cdot\vec{g}_{k_1})(\vec{d}_{31}\cdot\vec{g}_{k_2})(\vec{d}_{32}\cdot\vec{g}_{k_3})^2}{\hbar^4}\sum_{j,l=1}^{N}\int_0^t d\tau_1 e^{i(\omega_{k_1}+\omega_{k_2}-\omega_{21})\tau_1}\int_0^t d\tau_2 e^{-i(\omega_{k_2}+\omega_{k_3}-\omega_{21})\tau_2}$$
$$\times\frac{\langle U_{j1}^2(t-\tau_1)U_{l2}^1(t-\tau_2)\rangle(\omega_{31}+\omega_{32})^2}{(\omega_{32}+\omega_{k_3})^2(\omega_{31}-\omega_{k_3})^2}e^{i(\vec{k}_1-\vec{k}_2)\cdot\vec{r}}e^{-i(\vec{k}_2\cdot\vec{r}_l)+i(\vec{k}_1\cdot\vec{r}_j)}e^{i\vec{k}_3\cdot(\vec{r}_j-\vec{r}_l)}. \tag{19}$$

$$G_2(\vec{r},t)=\frac{1}{\hbar^4}\sum_{k_1\cdots k_4}(\vec{g}_{k_1}\cdot\vec{g}_{k_2})(\vec{g}_{k_3}\cdot\vec{g}_{k_4})\int_0^t d\tau_1 e^{i(\omega_{k_1}+\omega_{k_3}-\omega_{21})\tau_1}\int_0^t d\tau_2 e^{-i(\omega_{k_2}+\omega_{k_4}-\omega_{21})\tau_2}$$
$$\times\left(\frac{(\vec{d}_{32}\cdot\vec{g}_{k_1})(\vec{d}_{31}\cdot\vec{g}_{k_3})}{\omega_{k_3}-\omega_{31}}-\frac{(\vec{d}_{31}\cdot\vec{g}_{k_1})(\vec{d}_{32}\cdot\vec{g}_{k_3})}{\omega_{k_3}+\omega_{32}}\right)\left(\frac{(\vec{d}_{32}\cdot\vec{g}_{k_2})(\vec{d}_{31}\cdot\vec{g}_{k_4})}{\omega_{k_4}-\omega_{31}}-\frac{(\vec{d}_{31}\cdot\vec{g}_{k_2})(\vec{d}_{32}\cdot\vec{g}_{k_4})}{\omega_{k_4}+\omega_{32}}\right)$$
$$\times\sum_{j,l=1}^{N}e^{-i(\vec{k}_1+\vec{k}_3)\cdot(\vec{r}-\vec{r}_j)}e^{-i(\vec{k}_2+\vec{k}_4)\cdot(\vec{r}_l-\vec{r})}\langle U_{1j}^2(t-\tau_1)U_{2l}^1(t-\tau_2)\rangle. \tag{20}$$

The first- and second-order correlation functions for the EMF intensity are proportional to the similar correlation functions for the atomic subsystem $\langle U_{1j}^2(t-\tau_1)U_{2l}^1(t-\tau_2)\rangle$. Thus, after integrating over k_1, k_2, k_3, k_4, τ_1,τ_2 and using the vector identity

$$\sum_{\lambda_1\lambda_2}(\vec{e}_{\lambda_1}\cdot\vec{e}_{\lambda_2})(\vec{n}\cdot\vec{e}_{\lambda_1})(\vec{n}\cdot\vec{e}_{\lambda_2})=1-(\vec{k}_1\cdot\vec{n})^2-(\vec{k}_2\cdot\vec{n})^2+(\vec{k}_1\cdot\vec{k}_2)(\vec{k}_1\cdot\vec{n})(\vec{k}_2\cdot\vec{n}),$$

$$(\vec{n}\|\vec{d}_{3\alpha},\alpha=1,2)$$

one obtains the following expression for the $G_1(\vec{r},t)$, $G_2(\vec{r},t)$ correlation functions:

$$G_1(\vec{r},t)=\frac{d_{31}^2d_{32}^2}{\pi\hbar c^7}(\omega_{31}+\omega_{32})^2\sum_{j,l=1}^{N}\int_0^{\omega_{21}}dx\frac{x^3(\omega_{21}-x)^4}{(\omega_{32}+x)^2(\omega_{31}-x)^2}e^{(i/c)(\omega_{21}-x)(|\vec{r}+\vec{r}_l|-|\vec{r}+\vec{r}_j|)}\frac{(1-\cos^2\zeta)\chi(x,\vec{r}_{jl})}{|\vec{r}+\vec{r}_j||\vec{r}+\vec{r}_l|}$$
$$\times\left\langle U_{1j}^2\left(t-\frac{|\vec{r}+\vec{r}_l|}{c}\right)U_{2l}^1\left(t-\frac{|\vec{r}+\vec{r}_l|}{c}\right)\right\rangle\Theta\left(t-\frac{|\vec{r}+\vec{r}_j|}{c}\right)\Theta\left(t-\frac{|\vec{r}+\vec{r}_l|}{c}\right), \tag{21}$$

$$G_2(\vec{r},t)=\frac{d_{31}^2d_{32}^2(1-\cos^2\zeta_1)(1-\cos^2\zeta_2)}{4\pi c^8}(\omega_{31}+\omega_{32})^2\sum_{j,l=1}^{N}\left\{\int_0^{\omega_{21}}dx\frac{x^2(\omega_{21}-x)^2}{(x-\omega_{31})(x+\omega_{32})}\right\}^2$$
$$\times\frac{\cos(\omega_{21}|\vec{r}-\vec{r}_j|/c)\cos(\omega_{21}|\vec{r}-\vec{r}_l|/c)}{|\vec{r}-\vec{r}_j|^2|\vec{r}-\vec{r}_l|^2}\left\langle U_{1j}^2\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)U_{2l}^1\left(t-\frac{|\vec{r}-\vec{r}_l|}{c}\right)\right\rangle\Theta\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)\Theta\left(t-\frac{|\vec{r}-\vec{r}_l|}{c}\right). \tag{22}$$

Here $\Theta(t-|\vec{r}-\vec{r}_{j,l}|/c)$ is the Heaviside step function, and ζ is the angle between the direction of the vectors $\vec{r}$ and $\vec{d}_{3\alpha}$.

The retardation of the EMF in point $\vec{r}$ is taken into account in Eqs. (21) and (22). Markoff approximation allows us to ignore the retardation relative to the center of the atoms mass.

For the square intensity correlation function $G_4(\vec{r},t)$ we obtain the following expression:

$$G_4(\vec{r},t)=\frac{d_{31}^4d_{32}^4(1-\cos^2\zeta_1)^2(1-\cos^2\zeta_2)^2}{16\pi^2c^{16}}(\omega_{31}+\omega_{32})^4\left[\int_0^{\omega_{21}}\frac{x^2(\omega_{21}-x)^2dx}{(x-\omega_{31})(x+\omega_{32})}\right]^4\sum_{j,l=1}^{N}\frac{\cos(\omega_{21}|\vec{r}-\vec{r}_j|/c)\cos(\omega_{21}|\vec{r}-\vec{r}_l|/c)}{|\vec{r}-\vec{r}_j|^2|\vec{r}-\vec{r}_l|^2}$$
$$\times\sum_{m,n=1}^{N}\frac{\cos(\omega_{21}|\vec{r}-\vec{r}_m|/c)\cos(\omega_{21}|\vec{r}-\vec{r}_n|/c)}{|\vec{r}-\vec{r}_m|^2|\vec{r}-\vec{r}_n|^2}\left\langle U_{1j}^2\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)U_{1m}^2\left(t-\frac{|\vec{r}-\vec{r}_m|}{c}\right)U_{2n}^1\left(t-\frac{|\vec{r}-\vec{r}_n|}{c}\right)\right.$$
$$\left.\times U_{2l}^1\left(t-\frac{|\vec{r}-\vec{r}_l|}{c}\right)\right\rangle\Theta\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)\Theta\left(t-\frac{|\vec{r}-\vec{r}_l|}{c}\right)\Theta\left(t-\frac{|\vec{r}-\vec{r}_m|}{c}\right)\Theta\left(t-\frac{|\vec{r}-\vec{r}_n|}{c}\right). \tag{23}$$

Let us now express the correlation functions for the EMF intensity through the atomic subsystem correlation functions in the case of the cascade emission . At first, as the method of exclusion of the EMF operators in this case is not difficult, we get

$$G^{I}(\vec{r},t)=\sum_{k_1,k_2}(\vec{g}_{k_1}\cdot\vec{g}_{k_2})\frac{(\vec{d}_0\cdot\vec{g}_{k_1})(\vec{d}_0\cdot\vec{g}_{k_2})}{2\hbar^2}\sum_{j,l=1}^{N}e^{-i\vec{k}_2\cdot\vec{r}_l+i\vec{k}_1\cdot\vec{r}_j}\int_0^t d\tau_1 e^{i(\omega_{k_1}-\omega_0)\tau_1}\int_0^t d\tau_2 e^{-i(\omega_{k_2}-\omega_0)\tau_2}$$
$$\times\langle D_j^+(t-\tau_1)D_l^-(t-\tau_2)\rangle e^{-i(\vec{k}_1-\vec{k}_2\cdot\vec{r})}, \tag{24}$$

$$G^{II}(\vec{r},t)=\sum_{k_1\cdots k_4}(\vec{g}_{k_1}\cdot\vec{g}_{k_2})(\vec{g}_{k_3}\cdot\vec{g}_{k_4})\frac{(\vec{d}_0\cdot\vec{g}_{k_1})(\vec{d}_0\cdot\vec{g}_{k_2})(\vec{d}_0\cdot\vec{g}_{k_3})(\vec{d}_0\cdot\vec{g}_{k_4})}{4\hbar^4}\sum_{j,l,m,n=1}^{N}e^{-i\vec{k}_2\cdot\vec{r}_l+i\vec{k}_1\cdot\vec{r}_j-i\vec{k}_4\cdot\vec{r}_n+i\vec{k}_3\cdot\vec{r}_m}$$
$$\times\int_0^t d\tau_1 e^{i(\omega_{k_1}-\omega_0)\tau_1}\int_0^t d\tau_2 e^{-i(\omega_{k_2}-\omega_0)\tau_2}\int_0^t d\tau_3 e^{i(\omega_{k_3}-\omega_0)\tau_3}\int_0^t d\tau_4 e^{-i(\omega_{k_4}-\omega_0)\tau_4}\langle D_j^+(t-\tau_1)D_m^+(t-\tau_3)$$
$$\times D_n^-(t-\tau_4)D_l^-(t-\tau_2)\rangle e^{-i(\vec{k}_1+\vec{k}_3-\vec{k}_2-\vec{k}_4)\cdot\vec{r}}. \tag{25}$$

Here one can introduce the notations $G^{\mathrm{I}}(\vec{r},t)$ and $G^{\mathrm{II}}(\vec{r},t)$ for the first- and second-order correlation functions, respectively, of the two one-photon casade emission.

After summation over the polarization and integration over all directions of $\vec{k}$ we get for $G^{\mathrm{I}}(\vec{r},t)$,

$$G^{\mathrm{I}}(\vec{r},t)=\frac{d_0^2\omega_0^4}{2c^4}\sum_{j,l=1}^{N}\frac{e^{(i\omega_0/c)(|\vec{r}-\vec{r}_l|-|\vec{r}-\vec{r}_j|)}}{|\vec{r}-\vec{r}_l||\vec{r}-\vec{r}_j|}(1-\cos^2\zeta)\left\langle D_j^+\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)D_l^-\left(t-\frac{|\vec{r}-\vec{r}_l|}{c}\right)\right\rangle\Theta\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)\Theta\left(t-\frac{|\vec{r}-\vec{r}_l|}{c}\right) \tag{26}$$

and for $G^{\mathrm{II}}(\vec{r},t)$

$$G^{\mathrm{II}}(\vec{r},t)=\frac{d_0^4\omega_0^8}{4c^8}\sum_{j,l,m,n=1}^{N}\frac{e^{(i\omega_0/c)(|\vec{r}-\vec{r}_l|+|\vec{r}-\vec{r}_m|-|\vec{r}-\vec{r}_j|-|\vec{r}-\vec{r}_n|)}}{|\vec{r}-\vec{r}_j||\vec{r}-\vec{r}_l||\vec{r}-\vec{r}_m||\vec{r}-\vec{r}_n|}(1-\cos^2\zeta)^2\left\langle D_j^+\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)D_n^+\left(t-\frac{|\vec{r}-\vec{r}_n|}{c}\right)\right.$$
$$\left.\times D_l^-\left(t-\frac{|\vec{r}-\vec{r}_l|}{c}\right)D_m^-\left(t-\frac{|\vec{r}-\vec{r}_m|}{c}\right)\right\rangle\Theta\left(t-\frac{|\vec{r}-\vec{r}_j|}{c}\right)\Theta\left(t-\frac{|\vec{r}-\vec{r}_n|}{c}\right)\Theta\left(t-\frac{\vec{r}-\vec{r}_l|}{c}\right)\Theta\left(t-\frac{|\vec{r}-\vec{r}_m|}{c}\right). \tag{27}$$

Here the first- and second-order correlation functions are directly proportional to the emission rate and the square emission rate operators of the atomic subsystem, respectively. It is not difficult to observe that $G^{IV}(\vec{r},t)$ correlation function is proportional to the same order of the atomic correlation functions $D^+(t)D^-(t)$:

$$G^{\mathrm{IV}}(\vec{r},t)\sim\langle D_k^+(t_{rk})D_l^+(t_{rl})D_m^+(t_{rm})D_n^+(t_{rn})$$
$$\times D_i^-(t_{ri})D_j^-(t_{rj})D_f^-(t_{rf})D_s^-(t_{rs})\rangle.$$

Here $t_{ri}=t-|\vec{r}-\vec{r}_i|/c$ is the retardation time for the EMF generated by the ith atom.

It follows from Eqs. (21)–(27) that for two-photon dipole-forbidden transitions the second- and fourth-order correlation functions of the EMF are proportional to first- and second-order correlators of the $U_1^2(t)U_2^1(t)$ atomic operator product, respectively. For the two one-photon cascade emission the EMF correlation functions are of the order of the atomic correlation functions [see Eqs. (26), (27)]. In other words the product of two negative frequency parts of the EMF strength $E^-(t)E^-(t)$ in the correlation function for the EMF is proportional to the transition operator $U_1^2(t)$ in the case of two-photon dipole-forbidden transitions, while for the two one-photon cascade emission the negative frequency part of the EMF strength is proportional to the transition operators between the ground and excited state $D^+(t)$.

In the next section we shall analyze the time behavior of the functions $G_1(\vec{r},t)$, $G_2(\vec{r},t)$, $G_4(\vec{r},t)$, and $G^{\mathrm{I}}(\vec{r},t)$, $G^{\mathrm{II}}(\vec{r},t)$ for one and two atoms.

IV. THE COOPERATIVE BEHAVIOR OF THE ATOMIC SUBSYSTEM IN THE PROCESS OF TWO-PHOTON AND TWO ONE-PHOTON COOPERATIVE EMISSION

From the master equation (12a) it is easy to obtain the following chain of equations for two-photon dipole-forbidden cooperative transitions:

$$\frac{d}{dt}\langle R_{zj}(t)\rangle=-\sum_i F(i,j)[\langle R_i^+(t)R_j^-(t)\rangle+\langle R_j^+(t)R_i^-(t)\rangle], \tag{28}$$

$$\frac{d}{dt}\langle R_i^+(t)R_j^-(t)\rangle=2\sum_l[F(i,l)\langle R_l^+R_{zi}(t)R_j^-(t)\rangle+F(j,l)$$
$$\times\langle R_i^+(t)R_{zj}(t)R_l^-(t)\rangle],$$

and so on.

Here R_{zj}, R_j^+, and R_j^- are respectively the inversion, creation, and annihilation operators for the atomic subsystem

$$R_{zj}(t)=[U_{j2}^2(t)-U_{j1}^1(t)]/2,$$

$$R_j^+(t)=U_{j1}^2(t),R_j^-(t)=U_{j2}^1(t),$$

which satisfy the commutation relations

$$[R_j^+(t),R_i^-(t)]=2\delta_{ij}R_{zi}(t), \quad [R_{zj}(t),R_i^{\pm}(t)]=\pm\delta_{ij}R_i^{\pm}.$$

We are going to study the cooperative emission of two atoms situated at the distance r_{21}. It is possible to obtain a closed system of equations for two atoms at the second step of chain (28):

$$\frac{d}{dt}Z(t)=-\frac{1}{\tau_0}[1+Z(t)]-\frac{1}{\tau_1}V(t),$$

$$\frac{d}{dt}V(t)=-\frac{1}{\tau_0}V(t)+\frac{1}{\tau_1}Z(t)+\frac{4}{\tau_1}Y(t), \tag{29}$$

$$\frac{d}{dt}Y(t)=-\frac{2}{\tau_0}Y(t)-\frac{1}{2\tau_0}Z(t)+\frac{1}{2\tau_1}V(t).$$

Here we introduce variables: $Z(t)=\langle R_{z1}(t)\rangle+\langle R_{z2}(t)\rangle$ is the inversion of atoms, $V(t)=\langle R_2^+(t)R_1^-(t)\rangle+\langle R_1^+(t)R_2^-(t)\rangle$ is the cooperative rate of emission, $Y(t)=\langle R_{1z}(t)R_{2z}(t)\rangle$ is the correlation function of inversions for the first atom and the second one. The spontaneous and cooperative emission time τ_0, τ_1 can be found from the relations

$$\frac{1}{\tau_0}=\frac{8d_{31}^2d_{32}^2(\omega_{31}+\omega_{32})^2}{9\pi\hbar^2c^6}\int_0^{\omega_{21}}dx\,\frac{x^3(\omega_{21}-x)^3}{(\omega_{31}-x)^2(\omega_{32}+x)^2}$$

$$\frac{1}{\tau_1}=\frac{2d_{31}^2d_{32}^2(\omega_{31}+\omega_{32})^2}{\pi\hbar^2c^6}\int_0^{\omega_{21}}dx$$

$$\times\frac{x^3(\omega_{21}-x)^3\chi_{12}(x)\chi_{12}(\omega_{21}-x)}{(\omega_{32}+x)^2(\omega_{31}-x)^2}.$$

Using the initial conditions for variables, $Z(t=0)=1$, $V(t=0)=0$, $Y(t=0)=1/4$ we obtain the following solution of the system (29):

$$Z(\tau)=-\frac{4n^2}{1-n^2}e^{-2\tau}+\frac{1-n}{1+n}e^{-(1-n)\tau}+\frac{1+n}{1-n}e^{-(1+n)\tau}-1,$$

$$V(\tau)=-\frac{4n}{1-n^2}e^{-2\tau}-\frac{1-n}{1+n}e^{-(1-n)\tau}+\frac{1+n}{1-n}e^{-(1+n)\tau}, \tag{30}$$

$$Y(\tau)=\frac{1+n^2}{1-n^2}e^{-2\tau}-\frac{1-n}{2(1+n)}e^{-(1-n)\tau}$$

$$-\frac{1+n}{2(1-n)}e^{-(1+n)\tau}+1/4,$$

where $n=\tau_0/\tau_1$, $\tau=t/\tau_0$.

To estimate the dependence of the two-photon cooperative exchange integral between the atoms at a distance r_{12} we make the following approximation in the expression for the cooperative parameter n. First we average the exchange kernel $\chi_{12}(x)$ over the directions of the dipole momentum $\vec{d}$ and then one can approximate $x^2(\omega_{21}-x)^2\approx(\omega_{21}/2)^4$. In this case we obtain the following relation for parameter n:

$$n=3\left(\frac{c}{r_{12}\omega_{12}}\right)^2\left\{\frac{c}{r_{12}\omega_{12}}\sin\left(\frac{\omega_{21}r_{12}}{c}\right)-\cos\left(\frac{\omega_{21}r_{12}}{c}\right)\right\}.$$

We observe that at a big distance relative to parameter c/ω_{21} the exchange integral n oscillates as a function of distance r_{12} $[n\sim-\cos(\omega_{21}r_{12}/c)/r_{12}^2]$. In other words the two-photon cooperative exchange between the two atoms can inhibit or enhance the spontaneous emission as a function of the distance between the atoms and decreases as $1/r_{12}^2$.

Let us suppose that all the atoms are located within a volume whose linear dimensions are small compared to $\lambda_{\min}=2\pi c/\omega_{21}$. In this case $n\approx1$ and the solution of the system of equations (29) takes the form

$$Z(\tau)=2(1+\tau)e^{-2\tau}-1, \tag{31}$$

$$V(\tau)=2\tau e^{-2\tau},Y(\tau)=\tfrac{1}{4}(1-4\tau e^{-2\tau}).$$

Now we consider a more general case, when the number of atoms in the concentrated system is large. If we neglect the fluctuation of the number of excited atoms when the number of atoms $N\gg1$, we obtain from Eq. (28) the well-known Dicke equation for the atomic subsystem [7,12]:

$$\frac{d}{dt}Z(t)=-\frac{1}{\tau_0}[Z(t)+N/2]+\frac{1}{\tau_0}[Z^2(t)-N^2/4], \tag{32}$$

the solution of which is

$$Z(t)=-\frac{N}{2}th\frac{t-t_0}{2\tau_R},$$

where $t_0=\tau_R\ln N$ is the delay of the collective radiation pulse of the photon pair and $\tau_R=\tau_0/N$ is the collectivization time of the ensemble of atoms from the two-photon spontaneous decay of the $|2\rangle$ excited state.

The time behavior of one atom inversion in the process of the two one-photon cascade emission is similar to the cooperative emission of two atoms in the case of the dipole-

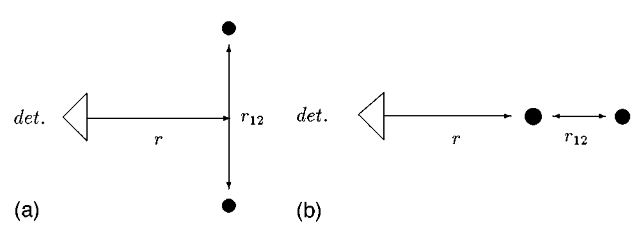

FIG. 2. (a) The detector is placed perpendicular to the axis between two atoms. (b) The detector is placed along to the axis between two atoms.

forbidden transition [see Eq. (31)]. Indeed, from master equation (15) we obtain the following system of equations for one atom:

$$\frac{d}{dt}\langle D_z(t)\rangle = -\frac{1}{2\tau_A}\langle D^+(t)D^-(t)\rangle,$$
$$\frac{d}{dt}\langle D^+(t)D^-(t)\rangle = \frac{2}{\tau_A}(1+\langle D_z(t)\rangle - \langle D^+(t)D^-(t)\rangle). \tag{33}$$

Taking into account the initial conditions, $\langle D_z(t=0)\rangle = 1$ and $\langle D^+(t=0)D^-(t=0)\rangle = 2$ we obtain the following time dependence of the atom variables

$$\langle D_z(t)\rangle = 2e^{-t/\tau_A} + \frac{t}{\tau_A}e^{-t/\tau_A} - 1$$
$$\langle D^+(t)D^-(t)\rangle = 2e^{-t/\tau_A} + \frac{2t}{\tau_A}e^{-t/\tau_A}. \tag{34}$$

Next we shall study the behavior of the fluctuations of the EMF intensity at a distance r from the center of mass of two atoms, excited relative to the dipole-forbidden transition.

In the case when the detector is situated perpendicular to the distance between the atoms [see Fig. 2(a)] we obtain the following dependence of the EMF fluctuations on time and distance $r_{12} = 2r_0$:

$$G_1(\vec{r},t) = \frac{d_{31}^2 d_{32}^2(\omega_{31}+\omega_{32})^2(1-\cos^2\zeta)}{\pi\hbar c^7|\vec{r}+\vec{r}_0|^2}\Theta^2(t_r)\int_0^{\omega_{21}}\frac{x^3(\omega_{21}-x)^4 dx}{(\omega_{32}+x)^2(\omega_{31}-x)^2}\{\tfrac{2}{3}[Z(t_r)+1]+\chi_{21}(x)V(t_r)\}, \tag{35}$$

$$G_2(\vec{r},t) = \frac{d_{31}^2 d_{32}^2(\omega_{31}+\omega_{32})^2(1-\cos^2\zeta)^2}{8\pi^2 c^8|\vec{r}-\vec{r}_0|^4}\Theta^2(t_r)\left(\int_0^{\omega_{21}}\frac{x^2(\omega_{21}-x)^2 dx}{(x-\omega_{31})(x+\omega_{32})}\right)^2[Z(t_r)+V(t_r)+1], \tag{36}$$

where $t_r = t - r/c$.

In the case when the detector is situated along the distance between the atoms [see Fig. 2(b)], the time dependence of the EMF correlation functions is

$$G_1(\vec{r},t) = \frac{d_{31}^2 d_{32}^2(\omega_{31}+\omega_{32})^2(1-\cos^2\zeta)}{\pi\hbar c^7|\vec{r}+\vec{r}_0|^2}\Theta^2(t_r)\int_0^{\omega_{21}}\frac{x^3(\omega_{21}-x)^4 dx}{(\omega_{32}+x)^2(\omega_{31}-x)^2}\{\tfrac{2}{3}[Z(t_r)+1]+\chi_{21}(x)\cos[2r_0(\omega_{21}-x)/c]V(t_r)\}, \tag{37}$$

$$G_2(\vec{r},t) = \frac{d_{31}^2 d_{32}^2(\omega_{31}+\omega_{32})^2(1-\cos^2\zeta)^2}{8\pi^2 c^8|\vec{r}-\vec{r}_0|^4}\Theta^2(t_r)\left(\int_0^{\omega\_21}\frac{x^2(\omega_{21}-x)^2 dx}{(x-\omega_{31})(x+\omega_{32})}\right)^2[Z(t_r)+\cos(2\omega_{21}r_0/c)V(t_r)+1]. \tag{38}$$

Here the time dependence of $Z(t_r)$ and $V(t_r)$ is given in Eqs. (31).

For the estimation of functions $G_1(\vec{r},t)$ and $G_2(\vec{r},t)$ we approximate $x^3(\omega_{21}-x)^4 \approx (\omega_{21}/2)^7$; $x^2(\omega_{21}-x)^2 \approx (\omega_{21}/2)^4$ and $\omega_{31}-x \approx \omega_{31}-\omega_{21}/2$; $\omega_{32}+x \approx \omega_{31}-\omega_{21}/2$. From Eqs. (35), (36) in this approximation we obtain

$$\Lambda_\perp = G_2(\vec{r},t) - [G_1(\vec{r},t)]^2 \sim \frac{3\hbar^2\omega_{21}^3\Theta^2(t_r)}{2^7\pi r^4 c^2\tau_0}\left[Z(t_r)+V(t_r)+1-\frac{8\Theta^2(t_r)\pi}{3\omega_{21}\tau_0}\left(Z(t_r)+1+\frac{3V(t_r)}{2\omega_{21}\tau_0}\int_0^{\omega_{21}}\chi_{21}(x)dx\right)^2\right].$$

A similar expression can be obtained for the case when the detector is situated along the vector r_{12}.

It follows from these relations that the second-order correlation function $G_2(\vec{r},t)$ is much greater than the first-order correlation function $G_1(\vec{r},t)$, because the parameter $(\tau_0\omega_{21})^{-1} \sim 10^{-7}$. The photons in the radiation field form time-correlated pairs. One can obtain $\Lambda_2(\vec{r},t)$ time dependence for a large number of excited atoms in the concentrated system.

$$\Lambda_2(\vec{r},t) = \frac{7\hbar^2\omega_{21}^3 N^2}{15\times 2^7\pi c^2 r^4\tau_0}\mathrm{sech}^2\left(\frac{t_r-t_0}{2\tau_R}\right) \times\left\{1-\frac{9N^2}{7\omega_{21}\tau_0}\mathrm{sech}^2\left(\frac{t_r-t_0}{2\tau_R}\right)\right\}.$$

However, one can observe that the second-order correlation function remains much greater than the square first-order correlation function, when the number of cooperative radiated photons increases. This superbunching phenomenon [18] may be distorted when the cooperative two-photon width (N^2/τ_0) of the levels achieves the value of ω_{21}. But this is impossible, because the dimension of the concentrated system of atoms is less than the minimum radiation wavelength $\lambda_{\min} = 2\pi c/\omega_{12}$. In this situation the number of atoms inside the volume V is less than $\lambda_{\min}^3/\rho^3$ (here ρ is the mean distance between the atoms). The superbunching phenomenon occurs in the two-photon dipole-forbidden superradiance because the photons in the coherence pairs (biphotons) belong to the broadband radiation field $\omega_k \in \omega_{21}$. We observe that at the point of observation $\vec{r}$ the density of the photon pairs is inversely proportional to the fourth power of the distance between the source and the detector.

It is very interesting to study the behavior of quantum fluctuations of the square intensity operator of the EMF:

$$\varepsilon_4 = \frac{G_4(\vec{r},t) - G_2^2(\vec{r},t)}{G_2^2(\vec{r},t)}. \tag{39}$$

When the distance from the system to the point of observation is much larger than the dimension of the atomic subsystem, and the dimension of the atomic subsystem is less than the radiation wavelength, the relative fluctuations of the square intensity correlation function is

$$\varepsilon_4 \approx \frac{\langle U_1^2(t_r)U_1^2(t_r)U_2^1(t_r)U_2^1(t_r)\rangle}{\langle U_1^2(t_r)U_2^1(t_r)\rangle^2} - 1. \tag{40}$$

From this expression it follows that $G_4(\vec{r},t)$ has the same value as $G_2^2(\vec{r},t)$ and using the solutions for the correlators (31) we obtain the following time dependence of ε_4 in the case of two atoms:

$$\varepsilon_4 = \frac{e^{2\tau}}{(1+2\tau)^2} - 1. \tag{41}$$

Here $\tau = t_r/\tau_0$.

From the equation for Λ and Eq. (41) it follows that the relative fluctuations of the EMF density operator $\varepsilon_2 = \{G_2(\vec{r},t) - G_1^2(\vec{r},t)\}/G_1^2(\vec{r},t)$ remain larger than unity with the increase in the number of atoms in the system, but the relative fluctuations of the square intensity correlation function is approximately equal to zero, because by increasing the number of atoms we can decouple the four particle correlator in the following manner:

$$\langle U_1^2(t_r)U_1^2(t_r)U_2^1(t_r)U_2^1(t_r)\rangle \approx \langle U_1^2(t_r)U_1^2(t_r)\rangle \times \langle U_2^1(t_r)U_2^1(t_r)\rangle.$$

In the second case, for the cascade emission we get

$$G^{\mathrm{I}}(\vec{r},t) = \frac{d_0^2\omega_0^4(1-\cos^2\zeta)}{c^4r^2}\left(\frac{t_r}{\tau_A}e^{-t_r/\tau_A} + e^{-t_r/\tau_A}\right)\Theta^2(t_r), \tag{42}$$

$$G^{\mathrm{II}}(\vec{r},t) = \frac{d_0^4\omega_0^8(1-\cos^2\zeta)^2}{c^8r^4}\Theta^2(t_r)e^{-t_r/\tau_A}. \tag{43}$$

We observe that for the cascade emission of two photons by a single atom the relative time fluctuation of the EMF density is

$$\varepsilon = \frac{G^{\mathrm{II}}(\vec{r},t) - [G^{\mathrm{I}}(\vec{r},t)]^2}{[G^{\mathrm{I}}(\vec{r},t)]^2} = \frac{e^{t_r/\tau_A}}{(t_r/\tau_A+1)^2} - 1$$

and it changes from a negative to a positive value in the time momentum $t_r = 2.513\tau_A$. In other words the fluctuation distribution changes from sub-Poisson to super-Poisson. Similar time behavior takes place for the relative fluctuations ε_4 in the case of two-photon dipole-forbidden transitions [see Eq. (41)]. For a large number of atoms $N \gg 1$ the first-order correlation function $[G^{\mathrm{I}}(\vec{r},t)]^2$ is approximately equal to the second-order correlation function $G^{\mathrm{II}}(\vec{r},t)$ and in this situation the two one-photon cooperative cascade emission is quasicoherent, $\varepsilon \approx 0$.

V. CONCLUSION

These results on the collective two-photon spontaneous decay for dipole-forbidden transitions may be applied to an extended system of atoms with low concentration. The two-atoms approximation can be taken into account here. As it follows from our results the two-photon interaction between radiators through virtual photon pairs changes significantly in extended media.

In the case of the two-photon dipole-forbidden transitions the product of the operators $E^+(t)E^+(t) \approx U_2^1(t)$ and $E^-(t)E^-(t) \approx U_1^2(t)$ represented the amplitude of the transitions between the excited and ground states, and the coherence appears only for these operators. In other words the relative fluctuations of the square intensity operator decrease with increasing the number of atoms $\varepsilon_4 \to 0$. The same behavior has the relative fluctuations of the EMF intensity operator in the two one-photon cascade emission.

APPENDIX

Below we shall show in what mode we get the relations (10) from (7) using the lemma and the relations (8). The exclusion of the free part for operators $C_{j3}^\dagger(t)$ and $C_{l3}(t-\tau)$ in the expression for I_1 can be obtained without using the lemma. If we substitute the relations (8) for $C_{j3}^\dagger(t)$ and $C_{l3}(t-\tau)$ operators in the correlator I_1 one obtains

$$I_1 = \sum_{kk_1k_2}\sum_{j,l=1}^{N}\sum_{\rho,\beta,\gamma,\delta=1}^{2}\frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)(\vec{d}_{3\gamma}\cdot\vec{g}_{k_1})(\vec{d}_{3\delta}\cdot\vec{g}_{k_2})}{\hbar^4}e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\int_0^t d\tau_1 e^{i\omega_3\tau_1}\int_0^{t-\tau}d\tau_2 e^{-i\omega_3\tau_2}$$
$$\times\langle[a_{k_1}(t-\tau_1)e^{i\vec{k}_1\cdot\vec{r}_j} - \mathrm{H.c.}]C_{j\gamma}^\dagger(t-\tau_1)[C_{j\beta}(t),O(t)]C_{l\rho}^\dagger(t-\tau)[a_{k_2}^\dagger(t-\tau_2)e^{-i(\vec{k}_2\cdot\vec{r}_j)} - \mathrm{H.c.}]C_{l\delta}(t-\tau_2)\rangle. \tag{A1}$$

If we decouple the operators of the EMF and take into account that $\langle a_{k_1}a_{k_2}^\dagger\rangle \approx \delta_{k_1k_2}$ in the Born-Markoff approximation we obtain the following expression from Eq. (A1):

$$I_1 \approx \sum_{k_1 k_2} \sum_{j,l=1}^{N} \frac{(\vec{d}_{32}\cdot\vec{g}_{k_1})^2(\vec{d}_{31}\cdot\vec{g}_{k_2})^2}{\hbar^4(\omega_{k_1}+\omega_{32})^2} e^{i(\vec{k}_1+\vec{k}_2)\cdot(\vec{r}_j-\vec{r}_l)}\zeta(\omega_{k_2}+\omega_{k_1}-\omega_{21})\langle C_{j2}^\dagger(t)[C_{j1}(t),O(t)]C_{l1}^\dagger(t)C_{l2}(t)\rangle. \tag{A2}$$

After the substitution of the free and source parts of operator $C_{j3}^\dagger$ in the expression for I_2 we get

$$I_2 = \sum_{kk_1} \sum_{j,l=1}^{N} \sum_{\rho,\beta,\gamma=1}^{2} \frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)(\vec{d}_{3\gamma}\cdot\vec{g}_{k_1})}{\hbar^3} e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\int_0^t d\tau_1 e^{i\omega_3\tau_1}\langle[a_{k_1}(t-\tau_1)e^{i\vec{k}_1\cdot\vec{r}_j}-\text{H.c.}]C_{j\gamma}^\dagger(t-\tau_1)$$
$$\times[C_{j\beta}(t),O(t)]C_{l3}^\dagger(t-\tau)C_{l\rho}(t-\tau)\rangle. \tag{A3}$$

Here the exclusion of the free part of the $C_{l3}^\dagger(t-\tau)$ operator is more difficult, because the free part of this operator is situated in the right-hand part of the I_2 correlator. In this case it is necessary to use the lemma. In what follows we purpose the notations

$$B(t-\tau_1)=\{a_{k_1}(t-\tau_1)e^{i\vec{k}_1\cdot\vec{r}_j}-\text{H.c.}\}C_{j\gamma}^\dagger(t-\tau_1),$$

$$D(t)=[C_{j\beta}(t),O(t)]$$

for the permutation of the free part of $C_{l3}^\dagger(t-\tau)$ operator from the right hand part to the left one.

After doing these notations we consecutively permute the free part of $C_{l3}^\dagger(t-\tau)$ according to the lemma

$$\begin{aligned}
&\langle B(t-\tau_1)D(t)C_{l3}^+(t-\tau)C_{l\rho}(t-\tau)\rangle\\
&=\langle B(t-\tau_1)D(t)\{C_{l3}^{s+}(t-\tau)+C_{l3}^{f+}(t-\tau)\}C_{l\rho}(t-\tau)\rangle\\
&=\langle B(t-\tau_1)D(t)\{C_{l3}^{s+}(t-\tau)+C_{l3}^{f+}(t)e^{-i\omega_3\tau}\}C_{l\rho}(t-\tau)\rangle\\
&=\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)\rangle+\langle B(t-\tau_1)D(t)\{C_{l3}^{+}(t)-C_{l3}^{s+}(t)\}C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&=\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)\rangle+\langle B(t-\tau_1)\{C_{l3}^{+}(t)D(t)-D(t)C_{l3}^{s+}(t)\}C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&=\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)\rangle+\langle B(t-\tau_1)\{C_{l3}^{f+}(t)+C_{l3}^{s+}(t)\}D(t)C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&\quad-\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t)C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&=\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)\rangle+\langle B(t-\tau_1)\{C_{l3}^{s+}(t)D(t)-D(t)C_{l3}^{s+}(t)\}C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&\quad+\langle B(t-\tau_1)C_{l3}^{f+}(t)D(t)C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&=\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)\rangle+\langle B(t-\tau_1)[C_{l3}^{s+}(t),D(t)]C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&\quad+\langle B(t-\tau_1)C_{l3}^{f+}(t-\tau_1)D(t)C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau+i\omega_3\tau_1}\\
&=\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)\rangle+\langle B(t-\tau_1)[C_{l3}^{s+}(t),D(t)]C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}+\langle B(t-\tau_1)\\
&\quad\times\{C_{l3}^{+}(t-\tau_1)-C_{l3}^{s+}(t-\tau_1)\}D(t)C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau+i\omega_3\tau_1}\\
&=\langle B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)\rangle+\langle B(t-\tau_1)[C_{l3}^{s+}(t),D(t)]C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau}\\
&\quad+\langle[C_{l3}^{s+}(t-\tau_1),B(t-\tau_1)]D(t)C_{l\rho}(t-\tau)\rangle e^{-i\omega_3\tau+i\omega_3\tau_1}
\end{aligned} \tag{A4}$$

Now we can write that

$$I_2 = \sum_{kk_1} \sum_{j,l=1}^{N} \sum_{\rho,\beta,\gamma=1}^{2} \frac{(\vec{d}_{3\beta}\cdot\vec{g}_k)(\vec{d}_{3\rho}\cdot\vec{g}_k)(\vec{d}_{3\gamma}\cdot\vec{g}_{k_1})}{\hbar^3} e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}\int_0^t d\tau e^{-i\omega_k\tau}\int_0^t d\tau_1 e^{i\omega_3\tau_1}\langle\{B(t-\tau_1)D(t)C_{l3}^{s+}(t-\tau)C_{l\rho}(t-\tau)$$
$$+B(t-\tau_1)[C_{l3}^{s+}(t),D(t)]C_{l\rho}(t-\tau)e^{-i\omega_3\tau}+[C_{l3}^{s+}(t-\tau_1),B(t-\tau_1)]D(t)C_{l\rho}(t-\tau)e^{-i\omega_3\tau+i\omega_3\tau_1}\}\rangle. \tag{A5}$$

After the Born-Markoff approximation and using the fact that $\langle a_{k_1}a_{k_2}^\dagger\rangle\approx\delta_{k_1k_2}$ it is not difficult to obtain the relation for I_2 from Eq. (10).

In a similar way [Eqs. (A1)–(A5)] we have obtained the expressions for I_3, I_4 correlators.

[1] M. Brune and J. M. Raimond, S. Haroche, Phys. Rev. A **35**, 154 (1987).

[2] M. Bruno and J. M. Raimond, P. Goy, Phys. Rev. Lett. **59**, 1898 (1987).

[3] Lin-Sheng He and Hun-Li Feng, Phys. Rev. A **49**, 4009 (1994).

[4] L. Gilles, B. M. Garreway, and P. L. Knight, Phys. Rev. A **49**, 2785 (1994).

[5] R. Ghosh, G. Y. Hong, Z. Y. Ou, and L. Mandel, Phys. Rev. A **34**, 3962 (1986).

[6] W. Lange, G. S. Agarwal, H. Walther, Phys. Rev. Lett. **76**, 3293 (1996).

[7] N. A. Enaki, Zh. Éksp. Teor. Fiz. **94**, 135 (1988) [Sov. Phys. JETP **67**, 2033 (1988)].

[8] N. A. Enaki and O. B. Prepelitsa, Teor. Mat. Fiz. **88**, 416 (1991).

[9] Zhidong Ghen and Helen Freedhoff, Phys. Rev. A **44**, 546 (1991).

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

[11] M. Gross and S. Haroche, Phys. Rep. **93**, 301 (1982).

[12] A. V. Andreev, V. I. Emel'yanov, and Yu. A. Il'inskiy, Usp. Fiz. Nauk. **131**, 653 (1980) [Sov. Phys. Usp. **23**, 493 (1980)].

[13] M. Ban, J. Opt. Soc. Am. B **10**, 1347 (1993).

[14] H. Carmichael, *An Open System Approach to Quantum Optics* (Springer-Verlag, Berlin, 1991).

[15] D. Klyshko, *Physical Foundation of Quantum Electronics* (in Russian) (Nauka, Moscow, 1986).

[16] G. S. Agarwal and S. Dutte Gupta, Phys. Rev. A **49**, 3954 (1994).

[17] C. W. Gardiner, *Quantum Noise* (Springer-Verlag, Berlin, 1990).

[18] J. Perina, *Quantum Statistics of Linear and Nonlinear Optical Phenomena* (Kluwer, Dordrecht, 1984).