Superradiation from two-photon spontaneous decay

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The present study carries out a theoretical investigation of the possibility of collectivization of an ensemble of atoms of a concentrated system from two-photon spontaneous decay. It is demonstrated that such a system of inverted atoms emits phase-correlated biphotons with an intensity proportional to the square of the number of atoms.

1. INTRODUCTION

Dicke's study[1] was the first to predict the possibility of collectivization of an ensemble of two-level atoms in one-photon spontaneous decay. This effect, which has come to be called superradiation, has become the focus of extensive theoretical and experimental investigation in this decade (see studies[2–4]). As demonstrated in Refs. 1–3, two-level atoms in an inverted quantum state are capable of collective light generation due to interaction through fluctuations in the electromagnetic field (EF) vacuum. In this case the rate of photon generation becomes proportional to N^2, where N in the number of atoms in the system.

The present study reports the possibililty of collectivization of an ensemble of atoms inverted with respect to the $|2\rangle - |1\rangle$ dipole-forbidden transition, where $|2\rangle$ is the first excited state; $|1\rangle$ is the ground state from the two-photon spontaneous decay of the $|2\rangle$ level. A concentrated system of atoms of dimensions substantially smaller than the minimum radiation wavelength is examined for simplicity. A photon pair whose total energy is fixed, $\hbar\omega_{k1} + \hbar\omega_{k2} = \hbar\omega_{21}$, is created in each decay event, where $\hbar\omega_{21}$ is the energy distance between the $|1\rangle$ and $|2\rangle$ levels. The common phases of such photon pairs may be in phase in spite of the fact that the photons in the pair have different energies. The radiation intensity of the photon pairs (biphotons) becomes proportional to N^2, as in one-photon superradiation. However the second-order photon correlation function remains much greater than the squared first-order correlation function during the entire collective decay process.

Section 2 develops a nonequilibrium technique for elimination of the two boson operators of the photons created in a single decay event. This technique is used to demonstrate that the atoms may get into phase in the spontaneous emission of biphotons, and will radiate with a rate of change in the population difference between the $|2\rangle$ and $|1\rangle$ levels proportional to N^2. Section 3 employs this technique to calculate the fluctuations in the radiation energy density at a distance of r from the radiation source.

The possibility of the photon subsystem acquiring coherent properties in two-photon processes was examined in Ref. 5–7. These studies consider the possibility of effective accumulation of coherent photons in a prescribed mode k_0, i.e., first order coherence. The present study investigates the possibility of the formation of phased photon pairs consisting of two quanta of different energies. In this respect the purpose of the study differs from that of Ref. 5–7.

2. KINETICS OF TWO-PHOTON DECAY

We will consider N three-level Λ-type radiators inverted with respect to the $|2\rangle$ and $|1\rangle$ levels. Since transitions between the $|2\rangle$ and $|3\rangle$, and $|1\rangle$ and $|3\rangle$ levels are allowed ($d_{32}, d_{31} \neq 0$), while a transition is forbidden between $|2\rangle$ and $|1\rangle$ ($d_{21} = 0$), the Hamiltonian of such a system takes the form

$$\hat{H} = \sum_{\alpha=1}^{3}\sum_{j=1}^{N} \hbar\omega_\alpha U_{j\alpha}{}^{\alpha} + \sum_k \hbar\omega_k a_k{}^+ a_k + i\sum_{\beta=1}^{2}\sum_k\sum_{j=1}^{N} \mathbf{d}_{3\beta}\mathbf{g}_k [a_k{}^+ C_j{}^*(\mathbf{k}) - \text{H.c.}](U_{j3}{}^{\beta} + U_{j\beta}{}^{3}). \tag{1}$$

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 $|\beta\rangle$ and $|3\rangle$;

$$C_j{}^*(\mathbf{k}) = \exp[-i\mathbf{k}\mathbf{r}_j], \quad \mathbf{g}_k = (2\pi\hbar\omega_k/V)^{1/2}\mathbf{e}_\delta,$$

$\mathbf{e}_\delta$ is the polarization vector of the photon ($\delta = 1,2$); V is the quantization volume; a_k^+ (a_k) is the Bose creation (destruction) operator of a photon with momentum $\hbar\mathbf{k}$, energy $\hbar\omega_k$ and polarization δ; $U_{j\alpha}^{\beta}$ is the operator of the transition between the α and β levels of the jth atom. In expression (1) and henceforth we will use Greek letters to describe summation over the atomic levels, and Latin letters for summation over the atomic sequence. The operators of the atomic subsystem and the electromagnetic field (EF) operators satisfy the commutation relations

$$[U_{l\beta}{}^{\alpha}, U_{j\beta'}^{\alpha'}] = \delta_{l,j}(\delta_{\beta,\alpha'}U_{j\beta'}^{\alpha} - \delta_{\alpha,\beta'}U_{j\beta}{}^{\alpha'}), \tag{2a}$$

$$[a_k, a_{k'}{}^+] = \delta_{k,k'}, \quad [a_k{}^+, a_{k'}{}^+] = [a_k, a_{k'}] = 0. \tag{2b}$$

The Heisenberg equations for the operators of the atomic subsystem and the EF obtained subject to (1) take the form

$$\frac{d}{dt}U_{j\alpha}{}^{\beta}(t) = i\omega_{\beta\alpha}U_{j\alpha}{}^{\beta} + \sum_k [a_k{}^+(t)C_j{}^*(\mathbf{k}) - \text{H.c.}] \cdot \left\{\frac{\mathbf{d}_{3\alpha}\mathbf{g}_k}{\hbar}U_{j3}{}^{\beta}(t) - \frac{\mathbf{d}_{3\beta}\mathbf{g}_k}{\hbar}U_{j\alpha}{}^{3}\right\}, \quad \alpha,\beta = 1,2, \tag{3a}$$

$$\frac{d}{dt}U_{j3}{}^{\beta}(t) = -i\omega_{3\beta}U_{j3}{}^{\beta} - \sum_k\sum_{\alpha=1}^{2}\frac{\mathbf{d}_{3\alpha}\mathbf{g}_k}{\hbar}[a_k{}^+(t)C_j{}^*(\mathbf{k}) - a_k(t)C_j(\mathbf{k})] \cdot [U_{j3}{}^{3}(t)\delta_{\alpha,\beta} - U_{j\alpha}{}^{\beta}(t)], \tag{3b}$$

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$$\frac{d}{dt}a_k(t)=-i\omega_k a_k(t)+\sum_{\alpha=1}^{2}\sum_{j=1}^{N}\frac{\mathbf{d}_{3\alpha}\mathbf{g}_k}{\hbar}C_j^*(\mathbf{k})[U_{j3}^{\alpha}(t)+U_{j\alpha}^{3}(t)]. \tag{3c}$$

The equations for $U_{j\beta}^{\alpha}$, $U_{j\alpha}^{3}$ and a_k^+ are hermitian-conjugate to the equations in (3a), (3b) and (3c), respectively.

Formally integrating equation (3c) we represent the operator $a_k(t)$ through the vacuum part a_k^v and the part related to the sources a_k^s:

$$a_k(t)=a_k^v(t)+a_k^s(t), \tag{4a}$$

$$a_k^s(t)=\sum_{\alpha=1}^{2}\sum_{j=1}^{N}\frac{\mathbf{d}_{3\alpha}\mathbf{g}_k}{\hbar}\int_0^t d\tau\, G_k(\tau)C_j^*(\mathbf{k})[U_{j3}^{\alpha}(t-\tau)+U_{j\alpha}^{3}(t-\tau)]d\tau,$$

$$G_k(\tau)=\exp(-i\omega_k\tau), \tag{4b}$$

where

$$a_k^v(t)|v\rangle=0,\quad \langle v|a_k^{v+}(t)=0,\quad a_k^v(t)=a_k^v(0)\exp[-i\omega_k t],$$

$|v\rangle$ is the wave is the wave function of the EF vacuum. After substitution of $a_k(t)$ and $a_k^+(t)$ into (3a) and averaging over the initial state of the "atom plus field" system $\psi=|v\rangle|A\rangle$ ($|A$ is the wave function of the atomic subsystem for $t=0$) we can easily obtain the following equation for the populations of the jth atom:

$$\frac{d}{dt}\langle U_{j\alpha}^{\alpha}(t)\rangle=-\sum_{l=1}^{N}\sum_{k}\sum_{\beta=1}^{2}\frac{(\mathbf{d}_{3\beta}\mathbf{g}_k)(\mathbf{d}_{3\alpha}\mathbf{g}_k)}{\hbar^2}\Big\{C_{jl}^*(\mathbf{k})\cdot\int_0^t d\tau\, G_k^*(\tau)\langle[U_{l\beta}^{3}(t-\tau)+U_{l3}^{\beta}(t-\tau)]\cdot[U_{j\alpha}^{3}(t)-U_{j3}^{\alpha}(t)]\rangle+\text{H.c.}\Big\}. \tag{5}$$

Since all atoms are in the $|2\rangle$ state at $t=0$, the one-photon transition to the $|1\rangle$ ground state is forbidden. This is easily determined by using the familiar Born-Markov approximation in the right side of (5).[2,3] Here the operators $U_{l3}^{\beta}(t-\tau)$ are replaced by $U_{l3}^{\beta}(t)\exp(i\omega_{3\beta}\tau)$, while the rightside of the equation (5) is equal to zero, since there are no spontaneous polarization sources in the diagonal part of the correlator $\psi_\alpha^\alpha(t,0)=\langle U_{l\alpha}^{3}(t)U_{j3}^{\alpha}(t)\rangle$ for $t=0$ (the diagonal part of this correlator is $\psi_\alpha^\alpha|_{l=j}=\langle U_{j3}^{3}(t)\rangle|_{t=0}=0$). Consequently the only path for the system to make the transition to the ground state is by two-photon decay through the intermediate $|3\rangle$ state. The Hamiltonian (1) as well as all the results obtained in the present article can easily be generalized to the case where the number of higher energy states is greater than unity. For this it is necessary to replace the index 3 by γ in all positions and to sum over γ. Below we will propose a more exact method of accounting for the delay in (5). This method allows incorporation of the two-photon decay diagrams in the collective radiation kinetics.

The following correlation functions are under the integral over τ in the right side of equation (5)

$$\psi_\alpha^{\beta}(t,\tau)=\langle U_{l3}^{\beta}(t-\tau)U_{j\alpha}^{3}(t)\rangle,$$

$$\psi^{\beta\alpha}(t,\tau)=\langle U_{l3}^{\beta}(t-\tau)U_{j3}^{\alpha}(t)\rangle, \tag{6a}$$

$$\psi_\beta^{\alpha}(t,\tau)=\langle U_{l\beta}^{3}(t-\tau)U_{j3}^{\alpha}(t)\rangle,$$

$$\psi_{\beta\alpha}(t,\tau)=\langle U_{l\beta}^{3}(t-\tau)U_{j\alpha}^{3}(t)\rangle. \tag{6b}$$

By simplifying the subsequent mathematical intermediate calculations we will consider only the equation for the functions $\psi_\alpha^\beta(t,\tau)$:

$$\frac{d}{d\tau}\psi_\alpha^{\beta}(t,\tau)=i\omega_{3\beta}\psi_\alpha^{\beta}(t,\tau)-\sum_{k}\sum_{\gamma=1}^{2}\frac{\mathbf{d}_{3\gamma}\mathbf{g}_k}{\hbar}\cdot\langle[a_k^+(t-\tau)C_l^*(\mathbf{k})-a_k(t-\tau)C_l(\mathbf{k})]\cdot[U_{l\gamma}^{\beta}(t-\tau)-\delta_{\gamma,\beta}U_{l3}^{3}(t-\tau)]U_{j\alpha}^{3}(t)\rangle. \tag{7}$$

The vacuum part of the operator $a_k^+(t-\tau)$ in equation (7) is easily eliminated. It is necessary to represent $a_k^+(t-\tau)$ in this case through the vacuum EF operator and the material operators in accordance with (4) and to operate with the operator $a_k^{v+}(t-\tau)$ on the bra vector $\langle\psi(0)|a_k^{v+}(t-\tau)=0$. It is more difficult to eliminate the vacuum part of the $a_k(t-\tau)$ operator. In this case it is necessary to transpose the operator to the right side of the correlator under the Sp sign. Since the $U_{j\alpha}^{3}(t)$ operator following $a_k(t-\tau)$ belongs to time t, its commutator with the vacuum part of the operator $a_k(t-\tau)$ and with the operator itself is nonzero. We will formulate the following lemma to eliminate the vacuum part of the EF operators in such situtations.

If the EF creation or destruction operator lies between the two operators of the atomic subsystem $\hat{A}(t_1)$ and $\hat{B}(t_2)$ belonging to other times, elimination of the vacuum part of this operator yields the following expression for the correlator:

$$\langle\hat{B}(t_2)\hat{a}_k(t)\hat{A}(t_1)\rangle=\langle\hat{B}(t_2)\hat{a}_s(t)\hat{A}(t_1)\rangle-\exp[-i\omega_k(t-t_1)]\langle\hat{B}(t_2)[\hat{a}_s(t_1),\hat{A}(t_1)]\rangle. \tag{8}$$

The proof of (8) can be obtained in the following manner. Since $a_k(t)=a_k^s(t)+a_k^v(t)$, we will represent the vacuum part through the vacuum-operator at time t_1 subject to the determination of $a_k^v(t)$ [see (4)]:

$$a_k^v(t)=a_k^v(t_1)\exp[-i\omega_k(t-t_1)]=[a_k(t_1)-a_k^s(t_1)]\exp[-i\omega_k(t-t_1)].$$

After substitution of $a_k^v(t)$ into the correlator we obtain

$$\langle\hat{B}(t_2)\hat{a}_k(t)\hat{A}(t_1)\rangle=\langle\hat{B}(t_2)\hat{a}_k^s(t)\hat{A}(t_1)\rangle+\exp[i\omega_k(t-t_1)]\langle\hat{B}(t_2)[\hat{a}_k(t_1)-a_k^s(t_1)]\hat{A}(t_1)\rangle,$$

$a_k(t_1)$ commutes with the operator $\hat{A}(t_1)$. Consequently taking into account that

$$a_k(t_1)A(t_1)|v\rangle=A(t_1)a_k^s(t_1)|v\rangle,$$

we easily obtain (8).

Subject to lemma (8) we obtain the following equation for $\psi_\alpha^\beta(t,\tau)$:

$$\frac{d}{d\tau}\psi_\alpha^{\beta}(t,\tau)=i\omega_{3\beta}\psi_\alpha^{\beta}(t,\tau)+V_\alpha^{\beta}(t,\tau), \tag{9}$$

$$V_\alpha^{\beta}(t,\tau)=\sum_{k}\sum_{n=1}^{N}\sum_{\gamma,\xi=1}^{2}\frac{(\mathbf{d}_{3\gamma}\mathbf{g}_k)(\mathbf{d}_{3\xi}\mathbf{g}_k)}{\hbar^2}$$

$$\cdot \int_0^{t-\tau} d\tau_1 \{-C_{ln}^*(\mathbf{k}) G_k^*(\tau_1) \langle [U_{n\gamma}^3(t-\tau-\tau_1)+\text{H.c.}]$$

$$\cdot [U_{l\xi}^\beta(t-\tau)-\delta_{\xi,\beta}U_{l3}^3(t-\tau)]U_{j\alpha}^3(t)\rangle + G_k(\tau_1)$$

$$\cdot C_{ln}(\mathbf{k})\langle[U_{l\xi}^\beta(t-\tau)-\delta_{\xi,\beta}U_{l3}^3(t-\tau)]\cdot$$

$$\cdot [U_{n3}^\gamma(t-\tau-\tau_1)+\text{э. c.}]U_{j\alpha}^3(t)\rangle\} + T_\alpha^\beta(t,\tau), \quad (9a)$$

$$T_\alpha^\beta(t,\tau) = -\sum_{n=1}^N\sum_k\sum_{\xi,\gamma=1}^2 \frac{(\mathbf{d}_{3\gamma}\mathbf{g}_k)(\mathbf{d}_{3\xi}\mathbf{g}_k)}{\hbar^2}\exp(i\omega_k\tau)$$

$$\cdot C_{ln}(\mathbf{k})\left\langle [U_{l\xi}^\beta(t-\tau)-\delta_{\xi,\beta}U_{l3}^3(t-\tau)]\left\{\int_0^t d\tau_1\, G_k(\tau_1)\right.\right.$$

$$\left.\left.\cdot[U_{n3}^\gamma(t-\tau_1)+U_{n\gamma}^3(t-\tau_1)],\quad U_{j\alpha}^3(t)\right\}_-\right\rangle,$$

$$C_{ln}(\mathbf{k}) = C_l(\mathbf{k})C_n^*(\mathbf{k}). \quad (9b)$$

Formally integrating equation (9) we obtain

$$\psi_\alpha^\beta(t,\tau) = \psi_\alpha^\beta(t,0)\exp(i\omega_{3\beta}\tau)$$

$$+\int_0^t d\tau_2\, V_\alpha^\beta(t,\tau_2)\exp[i\omega_{3\beta}(\tau-\tau_2)],$$

$$\psi_\alpha^\beta(t,0) = \langle U_{l3}^\beta(t)U_{i\alpha}^3(t)\rangle. \quad (10)$$

Analogous expressions like (9) and (10) are easily obtained for the remaining correlators (6). Substituting the expressions for these correlators into (5) we can obtain the following equation for the populations $\langle U_\alpha^\alpha(t)\rangle$:

$$\frac{d}{d\tau}\langle U_\alpha^\alpha(t)\rangle = -F_1^\alpha - F_2^\alpha, \quad \langle U_\alpha^\alpha(t)\rangle = \sum_{j=1}^N \langle U_{j\alpha}^\alpha(t)\rangle, \quad (11)$$

$$F_1^\alpha = \sum_{j,l=1}^N \sum_{k_1} \sum_{\beta=1}^2 \frac{(\mathbf{d}_{3\alpha}\mathbf{g}_{k_1})(\mathbf{d}_{3\beta}\mathbf{g}_{k_1})}{\hbar^2} \quad (11a)$$

$$\cdot\int_0^t d\tau \exp(i\omega_{k_1}\tau)C_{jl}^*(\mathbf{k}_1)[\exp(i\omega_{3\beta}\tau)\varphi_\alpha^\beta(t,0)$$

$$-\exp(-i\omega_{3\beta}\tau)\varphi_\beta^\alpha(t,0)] + \text{H.c.},$$

$$F_2^\alpha = \sum_{j,l=1}^N \sum_{k_1} \sum_{\beta=1}^2 \frac{(\mathbf{d}_{3\alpha}\mathbf{g}_{k_1})(\mathbf{d}_{3\beta}\mathbf{g}_{k_1})}{\hbar^2}$$

$$\cdot\int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2\, C_{jl}^*(\mathbf{k}_1)\exp(i\omega_{k_1}\tau_1)\{W_\alpha^\beta(t,\tau_2)$$

$$\cdot\exp[i\omega_{3\beta}(\tau_1-\tau_2)] - W_\beta^\alpha(t,\tau_2)\exp[-i\omega_{3\beta}(\tau_1-\tau_2)]\}+\text{H.c.} \quad (11b)$$

where

$$\varphi_\alpha^\beta(t,0) = \psi_\alpha^\beta(t,0) - \psi^{\beta\alpha}(t,0),$$

$$\varphi_\beta^\alpha(t,0) = \psi_\beta^\alpha(t,0) - \psi_{\beta\alpha}(t,0),$$

$$W_\alpha^\beta(t,\tau_2) = V_\alpha^\beta(t,\tau_2) - V^{\beta\alpha}(t,\tau_2),$$

$$W_\beta^\alpha(t,\tau_2) = V_\beta^\alpha(t,\tau_2) - V_{\beta\alpha}(t,\tau_2).$$

The first term in (11) takes into account the collective one-photon transition processes from the $|3\rangle$ level to the $|\alpha\rangle$ level ($\alpha = 1,2$). If the $|3\rangle$ level is populated, the primary contribution to (11a) comes from the correlators $\psi_\beta^\alpha(t,0) = \delta_{\alpha,\beta}\langle U_{j\alpha}^3(t)U_{l3}^\alpha(t)\rangle$. The remaining correlators rapidly oscillate in time with a frequency on the order of the transition frequencies in the system ω_{31}, ω_{32}. Since it was intially assumed that the $|3\rangle$ level is not populated, the contribution of this term to spontaneous two-photon decay is small compared to (11b). The term F_2^α is proportional to $g_k^2 g_{k2}^2$ (i.e., the second order term in the constant g_{k1}^2). In analyzing this term we will use the Markov approximation in the three-particle correlators from (9). For this purpose we represent the operators of the atomic subsystem under the integral signs over τ_1, τ_2, and τ_3 as

$$U_{l\gamma}^\beta(t-\tau_i) = \exp[i\omega_{\beta\gamma}(t-\tau_i)]\tilde{U}_{l\gamma}^\beta(t), \quad (12)$$

where $\tilde{U}_{l\gamma}^\beta(t)$ is a smooth function of time compared to $\exp(i\omega_{\beta\gamma}t)$. This approximation corresponds to neglecting the higher orders in the expansion in terms of the constant g_k^2 which couples with the EF vacuum and retaining only terms through second order.

In order to identify the more probable two-photon decay diagrams of the system from the $|2\rangle$ level we will integrate the coefficients of the three-particle correlators with respect to time after making approximation (12). Expressions (9) and (12) suggest that these three-particle correlators either have no spontaneous polarization sources, i.e., the diagonal part of such correlators vanishes when $l = j = n$, since the $|3\rangle$ level is not populated, or they have rapidly-oscillating multipliers of order $\exp(i\omega_{\alpha\beta}t)$. Hence they make not contribution to second order in the constant g_k^2. Clearly the terms obtained after commutation of the operators in (9b) make the primary contribution in this approximation:

$$T_\alpha^\beta(t,\tau) = -\sum_k \sum_{\xi,\gamma=1}^2 \frac{(\mathbf{d}_{3\gamma}\mathbf{g}_k)(\mathbf{d}_{3\xi}\mathbf{g}_k)}{\hbar^2}$$

$$\cdot\langle[\tilde{U}_{l\xi}^\beta(t)\exp[i\omega_{\beta\xi}(t-\tau)] - \delta_{\gamma,\xi}U_{l3}^3(t)]$$

$$\cdot[\tilde{U}_{j\alpha}^\gamma(t)\exp(i\omega_{\gamma\alpha}t) - \delta_{\gamma,\alpha}U_{j3}^3(t)]\rangle$$

$$\cdot\frac{\exp[-i(\omega_k-\omega_{3\gamma}-i0)t]-1}{-i(\omega_k-\omega_{3\gamma}-i0)}\exp(i\omega_k\tau). \quad (13)$$

Here

$$i/(x-i0) = iP/x - \pi\delta(x).$$

Substituting (14) and analogous expressions for $T^{\beta\alpha}$, $T_{\alpha\beta}$ and T_β^α into (12) we can easily determine that after integration with respect to τ_1 and τ_3 the two-particle correlators $\langle U_{j1}^2(t)U_{l2}^1(t)\rangle$ make the primary contribution to two-photon decay. The remaining two-particle correlators have rapidly oscillating time-dependent multipliers, and their contribution is insignificant on the average over the period $T = 2\pi/\omega_{21}$. In this approximation it is therefore possible to obtain the following expression for the populations of the system of atoms $\langle U_2^2\rangle$ and $\langle U_1^1\rangle$:

$$\frac{d}{dt}\langle U_1^1(t)\rangle = -\frac{d}{dt}\langle U_2^2(t)\rangle,$$

$$\frac{d}{dt}\langle U_2^2(t)\rangle=-4\pi\sum_{k_1,k_2}\sum_{j,l=1}^{N} C_{jl}^*(\mathbf{k}_1)C_{jl}(\mathbf{k}_2)$$

$$\cdot\delta(\omega_{k_1}+\omega_{k_2}-\omega_{21})\frac{(\mathbf{d}_{31}\mathbf{g}_{k_1})(\mathbf{d}_{32}\mathbf{g}_{k_2})}{\hbar^4}\langle U_{l1}^2(t)U_{j2}^1(t)\rangle$$

$$\cdot\left\{\frac{(\mathbf{d}_{31}\mathbf{g}_{k_2})(\mathbf{d}_{32}\mathbf{g}_{k_1})}{(\omega_{31}-\omega_{k_1})(\omega_{32}+\omega_{k_1})}+\frac{(\mathbf{d}_{31}\mathbf{g}_{k_1})(\mathbf{d}_{32}\mathbf{g}_{k_2})}{(\omega_{31}-\omega_{k_1})^2}\right\}.$$

For simplicity we will consider a concentrated system of atoms with dimensions much smaller than the minimum radiation wavelength. We have $C_{jl}^*(\mathbf{k}_1)=C_{jl}(\mathbf{k}_2)=1$ in this situation. As a result it is possible to go over to the collective operators

$$U_\nu^\lambda=\sum_{j=1}^{N}U_{j\nu}^\lambda,\quad [U_\nu^\lambda,U_{\nu'}^{\lambda'}]=U_{\nu'}^\lambda\delta_{\nu,\lambda'}-U_\nu^{\lambda'}\delta_{\nu',\lambda}.\quad(15)$$

in (1)–(14). After integration with respect to k_1 the following equation is easily obtain for $\langle U_2^2(t)\rangle$:

$$\frac{d}{dt}\langle U_2^2(t)\rangle=-\frac{1}{\tau_0}\langle U_1^2(t)U_2^1(t)\rangle,\qquad(16)$$

where

$$\frac{1}{\tau_0}=\frac{1}{\pi\tau_{31}\tau_{32}}\frac{\omega_{32}+\omega_{31}}{\omega_{31}^3\omega_{32}^3}\int_0^{\omega_{21}}dx\frac{x^3(\omega_{21}-x)^3}{(\omega_{31}-x)^2(\omega_{32}+x)},\qquad(17)$$

$$\tau_{3\beta}=3\hbar c^3/4\omega_{3\beta}^3 d_{3\beta}^2,\qquad \beta=1,2.$$

We then use the boson representation in equation (15) for the new collective operators

$$U_1^2=b_2^+b_1,\qquad U_2^1=b_1^+b_2,$$
$$U_2^2=b_2^+b_2,\qquad U_1'=b_1^+b_1.\qquad(18)$$

It is possible to obtain a closed system of equations for $\langle U_1^1\rangle$ and $\langle U_2^2\rangle$ by ignoring fluctuations of the operators U_1^1 and U_2^2, i.e., by decoupling the correlators $\langle U_1^2(t)U_2^1(t)\rangle$ in equations (14) and (16) in the following manner:

$$\langle b_2^+b_1b_1^+b_2\rangle=\langle b_2^+b_2\rangle(1+\langle b_1^+b_1\rangle).\qquad(18a)$$

Taking into account the fact that

$$\langle b_2^+b_2\rangle+\langle b_1^+b_1\rangle=N,\quad \langle b_2^+b_2\rangle-\langle b_1^+b_1\rangle=2R_{21},\qquad(19)$$

we obtain the following expressions for $\langle U_2^2\rangle$ and $\langle U_1^1\rangle$:

$$\langle U_2^2\rangle=R_{21}+N/2,\quad \langle U_1^1\rangle=N/2-R_{21}.\qquad(19a)$$

From (16)–(19) we therefore obtain the following expression for the population difference of the system:

$$\frac{d}{dt}R_{21}=-\frac{1}{\tau_0}\left(R_{21}+\frac{N}{2}\right)+\frac{1}{\tau_0}\left(R_{21}^2-\frac{N^2}{4}\right),\qquad(20)$$

whose solution is well-known in one-photon superradiation theory[1-3]:

$$R_{21}=-\frac{N}{2}\,\mathrm{th}\frac{t-t_0}{2\tau_R},\qquad(20a)$$

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 two-photon spontaneous decay of the $|2\rangle$ excited state. It follows from (20a) that the ensemble of atoms collectively emits photon pairs in the interval $\omega_k\in(0,\omega_{21})$ with a total energy $\hbar(\omega_{k1}+\omega_{k2})=\hbar\omega_{21}$. The rate of emission of such photon pairs is equal to

$$v_{21}=-\frac{dR_{21}}{dt}=\frac{N}{4\tau_R}\mathrm{sech}^2\frac{t-t_0}{2\tau_R}.\qquad(20b)$$

Decoupling of (18a) may also be obtained by breaking off the chain of equations for the two-particle correlators. By eliminating the bosonic operators of the photon subsystem using the scheme (5)–(14), we obtain the following equation for the two-particle correlator $\langle U_1^2(t)U_2^1(t)\rangle$:

$$\frac{d}{dt}\langle U_1^2(t)U_2^1(t)\rangle$$
$$=\frac{1}{\tau_0}\langle U_1^2(t)[U_2^2(t)-U_1^1(t)]U_2^1(t)\rangle.\qquad(21)$$

It follows from (16) and (21) that the chain of kinetic equations for the atomic subsystem in two-photon spontaneous decay is analogous to the chain of equations of superradiation in one-photon spontaneous decay.[1-3] We therefore decouple the right side of equation (21) in a semiclassical approximation in the following manner:

$$\langle U_1^2(U_2^2-U_1^1)U_2^1\rangle=-\langle U_1^2U_2^1\rangle+\langle U_1^2U_2'\rangle[\langle U_2^2\rangle-\langle U_1^1\rangle](1-1/N).\qquad(21a)$$

Thus equations (16) and (21) subject to (21a) form a system of kinetic equations for one-particle and two-particle correlators. The solution of this system agrees with the solution of (20a).

It is possible to obtain an equation for the change in photon density at ω_k using the boson operator elimination method discussed above. After the first elimination of the EF boson operators the equation for the photon density $n_k=\langle a_k^+a_k\rangle$ will take the form

$$\frac{d}{dt}n_k=\sum_{\beta,\alpha=1}^{2}\frac{(\mathbf{g}_k\mathbf{d}_{3\alpha})(\mathbf{g}_k\mathbf{d}_{3\beta})}{\hbar^2}\int_0^t d\tau\,G_k^*(\tau)$$
$$\cdot\langle[U_\alpha^3(t-\tau)+U_3^\alpha(t-\tau)][U_3^\beta(t)+U_\beta^3(t)]\rangle+\mathrm{H.c.}\quad(22a)$$

The number of photons radiated in the solid angle $\Delta\Omega$ and the spectral interval $\Delta\omega_k$ is determined from the relation

$$\mathcal{N}_k\Delta\Omega\,\Delta\omega_k=\frac{V}{(2\pi c)^3}n_k\omega_k^2\Delta\omega_k\Delta\Omega.$$

After the secondary elimination of the EF boson operators the equation for $\mathcal{N}_k$ takes the form

$$\frac{d}{dt}\mathcal{N}_k(t)=\frac{\omega_k^3(\omega_{21}-\omega_k)^3(\omega_{31}+\omega_{32})}{4\pi^2\tau_{31}\tau_{32}\omega_{31}^3\omega_{32}^3}\left\{\frac{1}{(\omega_k+\omega_{32})^2(\omega_{32}-\omega_k)}\right.$$
$$\left.+\frac{1}{(\omega_k-\omega_{31})^2(\omega_k+\omega_{32})}\right\}\langle U_2^2(t)\rangle(\langle U_1^1(t)\rangle+1).$$
$$(22b)$$

The number of photons of frequency ω_k emitted in direction $\mathbf{k}$ is directly proportional to the rate of light generation from two-photon collective decay, $v_{21}(t)$. The photons are created in pairs and the frequency dependence of the generation rate is determined primarily by the multiplier $\omega_k^3 (\omega_{21}-\omega_k)^3$. Equation (22b) is invariant under the substitution $\omega_k \to \omega_{21}-\omega_k$, while $\mathcal{N}_k$ adopts the greatest value for $\omega_k = \omega_{21}/2$. After integration of (22b) with respect to Ω_k and ω_k, we obtain $\mathcal{N}(t) = 2\langle U_1^1 \rangle$ for the total number of photons.

3. PHOTON CORRELATION

In recent years increasing attention has been devoted to the influence of photon statistics on the interaction of radiation with matter. Several studies in this area have been devoted to two-photon spontaneous radiation and light absorption.[8-10] Below we will consider the influence of collective photon pair radiation processes on EF density fluctuation as well as detection probabilities in two-photon spontaneous decay. Following Refs. 8, 11 we will consider the correlation functions

$$K_1(\mathbf{r},t)=\langle(\mathbf{E}^-(\mathbf{r},t)\mathbf{E}^+(\mathbf{r},t))\rangle, \tag{23a}$$

$$K_2(\mathbf{r},t)=\langle:(\mathbf{E}^-(\mathbf{r},t)\mathbf{E}^+(\mathbf{r},t))(\mathbf{E}^-(\mathbf{r},t)\mathbf{E}^+(\mathbf{r},t)):\rangle, \tag{23b}$$

where

$$\mathbf{E}^-(\mathbf{r},t)=\sum_k g_k a_k^+(t)e^{i\mathbf{kr}},$$

$:\hat{f}(r,t):$ indicates normal ordering, $K_1(\mathbf{r},t)$ is the EF density at the observation point $\mathbf{r}$, and $K_2(\mathbf{r},t)$ is the correlation function between the biphotons at the point $\mathbf{r}$. The function

$$\Lambda_2(\mathbf{r},t)=K_2(\mathbf{r},t)-K_1^2(\mathbf{r},t) \tag{23c}$$

takes into account the EF density fluctuation.

After partial elimination of the photon boson operators we obtain the following expressions:

$$K_1(\mathbf{r},t)=\sum_{k_1,k_2}(\mathbf{g}_{k_1}\mathbf{g}_{k_2})\sum_{\alpha,\beta=1}^{2}\frac{(\mathbf{d}_{3\alpha}\mathbf{g}_{k_1})(\mathbf{d}_{3\beta}\mathbf{g}_{k_2})}{\hbar^2}$$
$$\cdot\exp[-i(\mathbf{r},\mathbf{k}_1-\mathbf{k}_2)]\int_0^t d\tau_1\int_0^t d\tau_2\, G_{k_1}^*(\tau_1)G_{k_2}(\tau_2)\cdot$$
$$\cdot\langle[U_\alpha^3(t-\tau_1)+U_3^\alpha(t-\tau_1)][U_3^\beta(t-\tau_2)+U_\beta^3(t-\tau_2)]\rangle, \tag{24a}$$

$$K_2(\mathbf{r},t)=\sum_{k_1,k_4}\sum_{\alpha,\beta=1}^{2}(\mathbf{g}_{k_1}\mathbf{g}_{k_2})(\mathbf{g}_{k_3}\mathbf{g}_{k_4})\frac{(\mathbf{d}_{3\alpha}\mathbf{g}_{k_1})(\mathbf{d}_{3\beta}\mathbf{g}_{k_4})}{\hbar^2}$$
$$\cdot\int_0^t d\tau_1\int_0^t d\tau_4\, G_{k_1}^*(\tau_1)G_{k_4}(\tau_4)$$
$$\cdot\langle[U_\alpha^3(t-\tau_1)+U_3^\alpha(t-\tau_1)]a_{k_3}^+(t)a_{k_2}(t)[U_3^\beta(t-\tau_4)$$
$$+U_\beta^3(t-\tau_4)]\rangle$$
$$\cdot\exp[-i(\mathbf{r},\mathbf{k}_1+\mathbf{k}_3-\mathbf{k}_2-\mathbf{k}_4)]. \tag{24b}$$

After secondary elimination of the boson operators subject to (8) Eqs. (24) take the form

$$K_1(\mathbf{r},t)=\frac{2d_{31}^2d_{32}^2}{3\pi\hbar c^7r^2}(1-\cos^2\zeta)(\omega_{32}+\omega_{31})^2$$
$$\cdot\int_0^{\omega_{21}}\frac{x^3(\omega_{21}-x)^4\,dx}{(\omega_{32}+x)(\omega_{31}-x)}\langle U_1^2(t_r)U_2^1(t_r)\rangle\theta^2(t_r), \tag{25a}$$

$$K_2(\mathbf{r},t)=\frac{d_{31}^2d_{32}^2}{8c^8r^4\pi^2}(1-\cos^2\zeta)^2\left\{\int_0^{\omega_{21}}\frac{x^2(\omega_{21}-x)\,dx}{(\omega_{32}+x)(\omega_{31}-x)}\right\}^2$$
$$\cdot(\omega_{32}+\omega_{31})^2\langle U_1^2(t_r)U_2^1(t_r)\rangle\theta^2(t_r). \tag{25b}$$

Here $\theta(t_r)$ is the Heavyside step function, ζ is the angle between the direction of the vector $\mathbf{r}$ and $\mathbf{d}_{3\alpha}$, and $t_r = t - r/c$.

It follows from (25) that the second-order correlation function is much greater than the first-order correlation function $K_1(\mathbf{r},t)$. The photons in the radiation field form time-correlated pairs. This will cause an avalanche growth in EF density fluctuations at the point of observation $\mathbf{r}$. After substitution of (25) into (23c) and averaging over the direction of the dipoles $\mathbf{d}_{3\alpha}$ in the system of atoms we obtain the following expression:

$$\Lambda_2(\mathbf{r},t)=\frac{7\hbar^2\omega_{21}^3N^2}{15\cdot2^7\pi c^2r^4\tau_0}\operatorname{sech}^2\left(\frac{t_r-t_0}{2\tau_R}\right)$$
$$\cdot\left[1-\frac{9N^2}{7\omega_{21}\tau_0}\operatorname{sech}^2\left(\frac{t_r-t_0}{2\tau_R}\right)\right]\theta(t_r). \tag{26}$$

It was assumed in deriving (26) that $\omega_{31}, \omega_{32} \gg \omega_{21}$.

At the point of observation $\mathbf{r}$ the density of the photon pairs is inversely proportional to the fourth power of the distance from the source to the detector. It is possible to increase the biphoton density at the point of observation $\mathbf{r}$ by means of collecting lenses or mirrors.[10] In the present situation the biphoton detector is located at the image point of the source. We assume that the detector consists of the same two-level system with a dipole-forbidden transition between the $|1'\rangle$ ground state and the excited $|2'\rangle$ state. The effective Hamiltonian of the interaction of the detector with the source field takes the form

$$\hat{W}=\sum_{k_1k_2}(\hat{w}_{k_1k_2}^*a_{k_1}^+a_{k_2}^++\hat{w}_{k_1k_2}a_{k_1}a_{k_2}), \tag{27a}$$

where $\hat{\omega}_{k1k2}$ is the excitation matrix of the detector atoms. Obviously the probability of photon pair absorption is proportional to the squared matrix element:

$$\langle1'|W|2'\rangle^2\approx\frac{1}{2}\sum_{k_i(i=1,4)}w_{k_1k_3}^*w_{k_2k_4}\langle a_{k_3}^+a_{k_1}^+a_{k_2}a_{k_4}\rangle, \tag{27b}$$

i.e., the probability of photon pair absorption is proportional to the correlation function $K_2(\mathbf{r},t)$. As indicated by (26) and (27b) the phased photon pairs at the focal point of the lens or mirror act on the detector in a manner similar to that of coherent one-photon light on a two-level system with an allowed transition. The formation of phased photon pairs during collective spontaneous decay will serve to increase the photon pair detection rate. Unlike the two-photon spontaneous decay of individual atoms[10] in the present situation the rate of biphoton detection of time $t_r = t_0$ is proportional to the square of the number of atoms in the system, while the spontaneous decay time calculated per atom diminshes by a factor of N. When employing collecting lenses or mirrors to focus the biphotons the EF density at the focusing point diminishes by a factor of $r^2\Omega/s$, where s is the cross-sectional area of the focusing volume ($s^{1/2}\sim\omega_{21}c$), Ω is the solid angle of radiation focusing. In the present case the factor $1/r^4$ in (25) and (26) is replaced with Ω^2/s^2.

Experiments to determine the lifetime of the metastable states of hydrogen-and helium-like atoms in two-photon spontaneous decay with respect to the $2^2S_{1/2} \to 1^2S_{1/2}$ and $2^1S_0 \to 1^1S_0$ transitions have been discussed on several occasions in the literature.[12,13] Two-photon collective decay processes may also appear in such experiments. If a certain number of inverted atoms $N \sim 10$–100 arise at a distance less than the minimum radiation wavelength $\lambda_{\min} = hc/(E_{2S} - E_{1S}) = 1200\ [A]/Z^2$ (where $E_{2S} - E_{1S} = \hbar\omega_{21}$, Z is the ordinal number of the element), the lifetime of the $2S$ metastable state decreases: $t_a = t_0 = (\tau_0/N)\ln N$ (here $\tau_0 = 0.121Z^{-6}$ sec). In this case the exponential law of decay of the excited state changes substantially [see (20a)].

4. CONCLUSION

These results on collective two-photon spontaneous decay are applicable only to concentrated systems of atoms with dimensions less than the minimum radiation wavelength. In extended media the interaction between radiators through the virtual photon pairs changes significantly. The spatial separation of the photons in the pair will reduce the exchange interaction integral between atoms at a distance greater than the radiation wavelength [see (21)]. In spite of this fact the exchange interaction between atoms at a distance less than or on the order of the radiation wavelength may have a substantial influence on the two-photon spontaeous decay of extended media.[14] The latter must be taken into account in investigating collective two-photon processes in condensed media.

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