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DYNAMICS OF TWO-PHOTON PROCESSES IN TWO-LEVEL SYSTEM

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1. INTRODUCTION

In our previous papers /1-4/ the dynamics of a one-photon processes in a macroscopic two-level system was examined, and on the basis of Bogolubov's method /5/ and exact hierarhy was constructed. This hierarchy permits one to investigate a radiation intensity and some other characteristics of the superradiation process.

It is known, however, that multiphoton processes are realized in a two-level system, for a number of physically important cases. Such a situation occurs when there is a dipole-forbidden transition in the system. Another example is the so-called combination scattering process.

So, we shall here examine the kinetic problem for the case of two-photon processes in a macroscopic two-level system. Let such a system contain N two-level emitters in volume V_c, and each emitter is described by the operators: $r_j^{\pm} = (\sigma_{jx} \pm i\sigma_{jy})/2$, $r_{jz} = \sigma_{jz}/2$, where $\sigma_{j\alpha}$ is an α-th component of the Pauli spin operator and j is the "number" of an emitter in the system. It was shown in papers /6-9/ that the interaction of m photons with a two-level system for the case of a cascade-type process can be presented by the following operator

$$H_{int} = \hbar N^{-m/2} \sum_j g^{(m)} (r_j^- \prod_{f=1}^{m} a_f^+ + r_j^+ \prod_{f=1}^{m} a_f),$$

where a^+, a are photon operators and $g^{(m)}$ is a coupling constant. In a particular case of a two-photon-cascade-type process (fig.1) we shall examine an interaction of the form

$$H_{int}^{(c)} = \hbar N^{-1} \sum_j \sum_{\vec{k}}' \{ g_{\vec{k}} e^{i(\vec{k}+\vec{k}_b)\vec{x}_j} a_{\vec{k}} b r_j^+(\mu) + g_{\vec{k}}^* e^{-i(\vec{k}+\vec{k}_b)\vec{x}_j} a_{\vec{k}}^+ b^+ r_j^-(\mu) \}. \quad (1)$$

Here $\vec{x}_j$ is the radius-vector of j-th emitter, the photon mode $\vec{k}_b$ is singled out, the summation $\sum_{\vec{k}}'$ is extended to a complete set of modes with the exception of the mode $\vec{k}_b$ and $r_j^+(\mu) \equiv r_j^+ + \mu r_j^-$, $\mu \in \mathbf{R}$. The total Hamiltonian for the two-photon-cascade-type

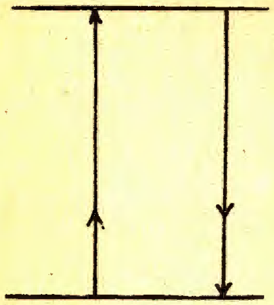
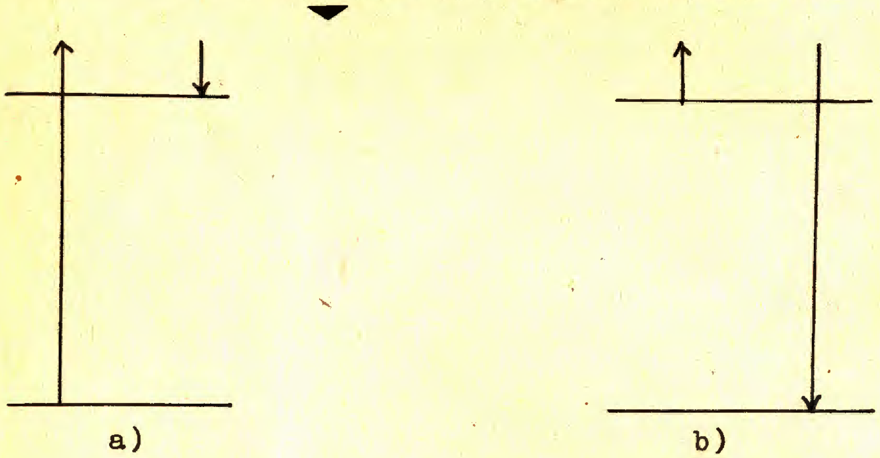


Fig.1. Cascade-type process in a two-level system.

Fig.2. Combination scattering process for the Stokes component radiation (a) and for the anti-Stokes component radiation (b).



process in the macroscopic two-level system is

$$H^{(c)} = H_F + H_E + H^{(c)}_{int} . \qquad (2)$$

Here H_F describes a free field $H_F = \sum_{\vec{k}} \hbar\omega_k a^+_{\vec{k}} a_{\vec{k}}$, and H_E is the energy of free emitters $H_E = \sum_j \hbar\Omega_j r_{jz}$, where $\hbar\Omega_j$ is the difference in level energies of j-th emitter.

For the combination scattering process the operator of the emitter-photon interaction has the form

$$H_{int} = \hbar N^{-(m+n)/2} \sum_j g^{(m,n)} \{ r^-_j \prod_{f=1}^{m} a^+_f \prod_{\ell=m+1}^{m+n} a_\ell + r^+_j \prod_{f=1}^{m} a_f \prod_{\ell=m+1}^{m+n} a^+_\ell \} .$$

In a particular case for the two-photon (Raman) scattering process, presented in fig.2a, we shall consider the following form of an interaction Hamiltonian

$$H^{(s)}_{int} = \hbar N^{-1} \sum_{j=1}^{N} \sum_{\vec{k}}{}' \{ g_{\vec{k}} e^{i(\vec{k}-\vec{k}_b)\vec{x}_j} a_{\vec{k}} b^+ r^-_j + g^*_{\vec{k}} e^{-i(\vec{k}-\vec{k}_b)\vec{x}_j} a^+_{\vec{k}} b r^+_j \} , \qquad (3)$$

where $\vec{k}_b$ is a wave-vector of a pumping mode for the Stokes-component radiation (fig.2a). Then the suitable total Hamiltonian is

$$H^{(s)} = H_F + H_E + H^{(s)}_{int} . \qquad (4)$$

The present paper has the following structure. In Sec.2 we begin by considering a model of the cascade-type process with Hamiltonian (2). An exact hierarchy will be derived by the method of partial elimination of the boson variables. The Markoffian equation for the two-photon cascade-type precess will be examined in Sec.3. Results for the emission and absorption processes are obtained here by solving a radiation equation. Sect.4 will be devoted to the construction of hierarchy for the combination scattering process with Hamiltonian (4). The spontaneous radiation of the Stokes component and its angular distribution will be examined. Here we shall consider also the induced Stokes radiation together with the pumping amplification effect. Qualitative discussions and conclusive remarks will be given in the final section.

2. EXACT HIERARCHY FOR A CASCADE-TYPE SYSTEM (2)

Let us present Hamiltonian (2) in the following manner

$$H^{(c)} = H_{\tilde{F}} + H_{\tilde{E}} + H^{(c)}_{int} , \qquad (5)$$

where $H_{\tilde{F}} = \sum_{\vec{k}}{}' \hbar\omega_k a^+_{\vec{k}} a_{\vec{k}}$, $H_{\tilde{E}} = H_E + \hbar\omega_b b^+ b$.

Let D_t be the statistical operator for system (5). Then

$$i\hbar \frac{\partial}{\partial t} D_t = [H^{(c)}, D_t] , \qquad (6)$$

and we shall consider the following form of initial conditions

$$D_{t_0} = D(\tilde{E}) \otimes D(\tilde{F}) , \qquad (7)$$

where $D(\tilde{E})$ is the statistical operator for free emitters and the pumping mode, $D(\tilde{F})$ is the one for a free field in equilibrium: $D(\tilde{F}) = \exp(-\beta H_{\tilde{F}}) / Tr_{(\tilde{F})} \exp(-\beta H_{\tilde{F}})$.

Let is define by $\mathcal{C}$ an arbitrary operator acting on the variables only of $\tilde{E}$-subsystem. It can be $r_{j\alpha}$, b^+, b or some combination of these operators. The equation of motion for $\mathcal{C}$ in the Heisenberg representation is

$$i\hbar \frac{\partial}{\partial t} \mathcal{O}(t) = [\mathcal{O}(t), H^{(e)}]. \qquad (8)$$

For the dynamical average we have /1,4,5/

$$<\mathcal{O}(t)> = \mathop{\mathrm{Tr}}_{(E,F)} \mathcal{O} D_t = \mathop{\mathrm{Tr}}_{(E,F)} \mathcal{O}(t) D_{t_0} . \qquad (9)$$

The equation of motion for the variables $a_{\vec k}$ is

$$i\hbar \frac{\partial}{\partial t} a_{\vec k} = \hbar\omega_k a_{\vec k} + \hbar N^{-1}\sum_j g^*_{\vec k} e^{-i(\vec k+\vec k_b)\vec x_j} b^+ r_j^-(\mu),$$

and its formal solution can be represented in the form

$$a_{\vec k}(t) = a_{\vec k}(t_0) e^{-i\omega_k(t-t_0)} -$$

$$- iN^{-1} g^*_{\vec k}\sum_j e^{-i(\vec k+\vec k_b)\vec x_j}\int_{t_0}^{t} e^{-i\omega_k(t-\tau)} b^+(\tau) r_j^-(\mu,\tau)\,d\tau.$$

Substituting now $a_{\vec k}(t)$ and $a^+_{\vec k}(t)$ into equation (8), averaging, and eliminating the variables $a^+_{\vec k}(t_0)$, $a_{\vec k}(t_0)$ in compliance with Bogolubov's methos /6/ (see also /1,4/) one can obtain in the limit $t_0\to-\infty$ that

$$\frac{d}{dt}<\mathcal{O}(t)> + \frac{i}{\hbar}<[\mathcal{O}(t), H_E(t)]> =$$

$$= \sum_{\vec\nu,\vec\nu'} \int_{-\infty}^{t} d\tau \{ D(\vec\nu,\vec\nu';t,\tau) <[R^+_{\vec\nu}(t)\, b(t), \mathcal{O}(t)]\, R^-_{\vec\nu'}(\tau)\, b^+(\tau)> +$$

$$+ N(\vec\nu,\vec\nu';t,\tau)<[[R^+_{\vec\nu}(t)\, b(t),\mathcal{O}(t)], R^-_{\vec\nu'}(\tau)\, b^+(\tau)]> +$$

$$+\mu^2 D(-\vec\nu,-\vec\nu';t,\tau)<[R^-_{\vec\nu}(t)\, b(t),\mathcal{O}(t)]R^+_{\vec\nu'}(\tau)\, b^+(\tau)> +$$

$$+\mu^2 N(-\vec\nu,-\vec\nu';t,\tau)<[[R^-_{\vec\nu}(t)\, b(t),\mathcal{O}(t)], R^+_{\vec\nu'}(\tau)\, b^+(\tau)]> +$$

$$+\mu D(\vec\nu,-\vec\nu';t,\tau)<[R^+_{\vec\nu}(t)\, b(t),\mathcal{O}(t)]\, R^+_{\vec\nu'}(\tau)\, b^+(\tau)> +$$

$$+\mu N(\vec\nu,-\vec\nu';t,\tau)<[[R^+_{\vec\nu}(t)\, b(t),\mathcal{O}(t)], R^+_{\vec\nu'}(\tau)\, b^+(\tau)]> +$$

$$+\mu D(-\vec\nu,\vec\nu';t,\tau)<[R^-_{\vec\nu}(t)\, b(t),\mathcal{O}(t)]\, R^-_{\vec\nu'}(\tau)\, b^+(\tau)> +$$

$$+\mu N(-\vec\nu,\vec\nu';t,\tau)<[[R^-_{\vec\nu}(t)\, b(t),\mathcal{O}(t)], R^-_{\vec\nu'}(\tau)\, b^+(\tau)]> \} + h.c. \qquad (10)$$

Here $R^\pm_{\vec\nu}$ is a collective operator $R^\pm_{\vec\nu} \equiv \sum_j r_j^\pm e^{\pm i\vec\nu\vec x_j}$,

and $\{\vec\nu\}$ corresponds to N modes on the volume V_c $\vec\nu=(\nu_\alpha)$, $\nu_\alpha = \frac{2\pi n_\alpha}{L_\alpha}$, $\alpha = x, y, z$, $n = 1, 2, \dots$, $V_c=\prod_\alpha L_\alpha$. The functions D(·) and N(·) are

$$D(\vec\nu,\vec\nu';t,\tau) \equiv N^{-2}\sum_{\vec k}|g_{\vec k}|^2 \phi(\vec k+\vec k_b-\vec\nu)\phi^*(\vec k+\vec k_b-\vec\nu')e^{-i\omega_k(t-\tau)},$$

$$N(\vec\nu,\vec\nu';t,\tau) \equiv N^{-2}\sum_{\vec k}|g_{\vec k}|^2 N_k \phi(\vec k+\vec k_b-\vec\nu)\phi^*(\vec k+\vec k_b-\vec\nu')e^{-i\omega_k(t-\tau)},$$

where $\phi(\vec\kappa) \equiv N^{-1}\sum_j e^{i\vec\kappa\vec x_j}$, $N_k \equiv \exp(-\frac{1}{2}\beta\hbar\omega_k)/\,2\sinh(\frac{1}{2}\beta\hbar\omega_k)$.

Expression (10) is an exact hierarchy of the kinetic equations for the $\tilde E$-subsystem in the cascade-type system with Hamiltonian (5)

If there is only weak interaction between the emitters and field in (1), one can use the following representation (zero approximation)

$$R^\pm_{\vec\nu}(\tau) \cong R^\pm_{\vec\nu}(t)\, e^{\mp i\Omega(t-\tau)}\, e^{-(t-\tau)/2T}, \quad b(\tau) \cong b(t)\, e^{i\omega_b(t-\tau)} ,$$

where Ω and T are parameters of the Lorentz inhomogeneous broadening /10/: Ω is a central frequency in the Lorentz distribution and T is the life-time of oscillators. Now ignoring in (10) the contribution from high-frequency oscillating collective terms R^+R^+, R^-R^- and from factors D(·) , N(·) at $\vec\nu\neq\vec\nu'$ one can obtain

$$\frac{d}{dt}<\mathcal{O}(t)> + \frac{i}{\hbar}<[\mathcal{O}(t), H_{\tilde E}(t)]> =$$

$$= \frac{1}{2N}\sum_{\vec\nu}\{\Gamma^-_{\vec\nu}<[R^+_{\vec\nu}(t)\, b(t), \mathcal{O}(t)]R^-_{\vec\nu}(t)\, b^+(t)> +$$

$$+ N^-_{\vec\nu}<[[R^+_{\vec\nu}(t)\, b(t),\mathcal{O}(t)], R^-_{\vec\nu}(t)\, b^+(t)]> + \mu^2\Gamma^+_{\vec\nu}<[R^-_{\vec\nu}(t)\, b(t),\mathcal{O}(t)]R^+_{\vec\nu}(t)\, b^+(t)> + \qquad (11)$$

$$+ \mu^2 N^+_{\vec\nu}<[[R^-_{\vec\nu}(t)\, b(t),\mathcal{O}(t)], R^+_{\vec\nu}(t)\, b^+(t)]> + h.c.\}.$$

Here $\Gamma^\pm_{\vec\nu} \equiv \gamma^\pm_{\vec\nu} - iQ^\pm_{\vec\nu}$,

$$\gamma^\pm_{\vec\nu} \equiv \frac{4TV}{(2\pi)^3 N}\int d\vec k\, \frac{|g_{\vec k}\phi(\vec k+\vec k_b\pm\vec\nu)|^2}{1+4T^2(\omega_k+\omega_b\pm\Omega)^2},$$

$$Q^\pm_{\vec\nu} \equiv \frac{8T^2V}{(2\pi)^3 N}\int d\vec k\, |g_{\vec k}\phi(\vec k+\vec k_b\pm\vec\nu)|^2\, \frac{\omega_k+\omega_b\pm\Omega}{1+4T^2(\omega_k+\omega_b\pm\Omega)^2},$$

$$N^{\pm}_{\vec\nu} \equiv n^{\pm}_{\vec\nu} - i\kappa^{\pm}_{\vec\nu},$$

$$n^{\pm}_{\vec\nu} \equiv \frac{4TV}{(2\pi)^3 N}\int d\vec{k}\, N_k \frac{|g_{\vec k}\phi(\vec k + \vec k_b \pm \vec\nu)|^2}{1+4T^2(\omega_k+\omega_b\pm\Omega)^2},$$

$$\kappa^{\pm}_{\vec\nu} \equiv \frac{8T^2V}{(2\pi)^3 N}\int d\vec{k}\, N_k |g_{\vec k}\phi(\vec k + \vec k_b \pm \vec\nu)|^2 \frac{\omega_k+\omega_b\pm\Omega}{1+4T^2(\omega_k+\omega_b\pm\Omega)^2}.$$

One can consider equation (11) as a Markoffian one for the two-photon-cascade-type process in the macroscopic two-level system (5). Below we shall examine some consequences from (11)

3. RADIATION AND ABSORPTION PROCESSES IN THE SYSTEM (5)

At first let us consider the simplest case of $\mu = 0$ (the rotating wave approximation [10]). Then, due to the commutation rules $[R_z, R^{\pm}_{\vec\nu}] = \pm R^{\pm}_{\vec\nu}$, $[R^{+}_{\vec\nu}, R^{-}_{\vec\nu}] = 2R_z$. For the case of $\mathcal{O}(t) = R_z(t)$ from (11) one can obtain

$$\frac{d}{dt}\langle R_z\rangle = -N^{-1}\sum_{\vec\nu}\{\gamma^{-}_{\vec\nu}\langle R^{+}_{\vec\nu}R^{-}_{\vec\nu}bb^{+}\rangle + n^{-}_{\vec\nu}\langle 2R_z b^{+}b + R^{+}_{\vec\nu}R^{-}_{\vec\nu}\rangle\}. \qquad (12)$$

Further for $\mathcal{O}(t) = b^{+}(t)\,b(t)$ we get

$$\frac{d}{dt}\langle b^{+}(t)b(t)\rangle = -\frac{d}{dt}\langle R_z(t)\rangle. \qquad (13)$$

It is known that the radiation intensity can be defined as [10]

$$I(t) = -\hbar\Omega\frac{d}{dt}\langle R_z(t)\rangle. \qquad (14)$$

When $I(t) > 0$, the radiation process takes place in the system. On the contrary, for $I(t) < 0$ there is an absorption in the system.

Let us consider some new operator

$$S_{\vec\nu} = \sum_{\substack{j,j'\\(j\neq j')}} r^{+}_{j} r^{-}_{j'} e^{i\vec\nu(\vec x_j - \vec x_{j'})}.$$

Then $R^{\pm}_{\vec\nu}R^{\mp}_{\vec\nu} = S_{\vec\nu} + N/2 \pm R_z$.

Now from equation (12) we obtain

$$\frac{d}{dt}\langle R_z(t)\rangle = -N^{-1}\sum_{\vec\nu}\{\langle(S_{\vec\nu}+\frac{N}{2})(\gamma^{-}_{\vec\nu}bb^{+}+n^{-}_{\vec\nu})\rangle + (\gamma^{-}_{\vec\nu}+2n^{-}_{\vec\nu})\langle R_z bb^{+}\rangle - n^{-}_{\vec\nu}\langle R_z\rangle\}. \qquad (15)$$

From expression (13) we have that $\langle R_z(t)\rangle + \langle b^{+}(t)\,b(t)\rangle = \text{const} \equiv M$ where M is defined by the initial conditions (7). Let us suppose the correlations to be weak between the emitters and coherent pumping field. Then

$$\langle S_{\vec\nu}bb^{+}\rangle \cong \langle S_{\vec\nu}\rangle\langle bb^{+}\rangle = \langle S_{\vec\nu}\rangle(M+1-\langle R_z\rangle),$$

$$\langle R_z bb^{+}\rangle \cong \langle R_z\rangle\langle bb^{+}\rangle = \langle R_z\rangle(M+1-\langle R_z\rangle).$$

If at the initial time moment all emitters are excited in the so-called Bloch state [10], then

$$\langle S_{\vec\nu}\rangle = \{\Gamma(\vec\nu-\vec\nu_0) - N^{-1}\}(N^2/4 - \langle R_z\rangle^2),$$

where $\Gamma(\vec\nu-\vec\nu_0) \equiv |N^{-1}\sum_j \exp\{i(\nu-\nu_0)x_j\}|^2$, and vector ν_0 describes an initial excited state of the emitter subsystem. So, from (15) we obtain

$$\frac{d}{dt}\langle R_z\rangle = -N^{-1}\sum_{\vec\nu}\{\Gamma(\vec\nu-\vec\nu_0) - N^{-1}\}(\frac{N^2}{4}-\langle R_z\rangle^2)\}\{\gamma^{-}_{\vec\nu}(M+1-\langle R_z\rangle)+n^{-}_{\vec\nu}\} - \frac{1}{2}\gamma(M+1-\langle R_z\rangle) - \frac{n}{2} - N^{-1}(\gamma+2n)\langle R_z\rangle(M+1-\langle R_z\rangle)\} + \frac{n}{N}\langle R_z\rangle, \qquad (16)$$

where $\gamma \equiv \sum_{\vec\nu}\gamma^{-}_{\vec\nu}$, $n \equiv \sum_{\vec\nu} n^{-}_{\vec\nu}$. To integrate equation (16), one ought to determine roots of the polynomial of the third degree in its right-hand side. For simplicity let us consider the case of zero temperature $\beta^{-1} = 0$ and an initial state only with the pumping mode $\vec k_b$ of the field. Then, instead of (16) we have

$$\frac{d}{dt}\langle R_z\rangle = -\frac{\alpha}{N}(\langle R_z\rangle - M - 1)(\langle R_z\rangle + \frac{N}{2})(\langle R_z\rangle - \frac{N}{2} - \frac{\gamma}{\alpha}), \qquad (17)$$

where $\alpha \equiv \sum_{\vec\nu}\gamma^{-}_{\vec\nu}\{\Gamma(\vec\nu-\vec\nu_0) - N^{-1}\} \leq \gamma$.

It is obvious that

$$\{(\langle R_z\rangle - M - 1)(\langle R_z\rangle + \frac{N}{2})(\langle R_z\rangle - \frac{N}{2} - \frac{\gamma}{\alpha})\}^{-1} = \frac{A}{\langle R_z\rangle - M - 1} + \frac{B}{\langle R_z\rangle + \frac{N}{2}} + \frac{C}{\langle R_z\rangle - \frac{N}{2} - \frac{\gamma}{\alpha}},$$

where $A^{-1} \equiv (M+1+\frac{N}{2})(M+1-\frac{N}{2}-\frac{\gamma}{\alpha})$, $B^{-1} \equiv (N+\frac{\gamma}{\alpha})(M+1+\frac{N}{2})$,

$C^{-1} \equiv (N+\frac{\gamma}{\alpha})(\frac{N}{2}+\frac{\gamma}{\alpha}-M-1)$.

The solution of (17) can be now represented in the following form

$$e^{-\alpha(t-\tilde{t}_0)/N} = |\langle R_z\rangle - M - 1|^A \cdot |\langle R_z\rangle + \frac{N}{2}|^B \cdot |\langle R_z\rangle - \frac{N}{2} - \frac{\gamma}{\alpha}|^C . \quad (18)$$

Here the value of $\tilde{t}_0$ can be determined from the initial value of $\langle R_z\rangle$. One can easily see that $A + B + C = 0$. Let us choose $M = \frac{N}{2} + \frac{\gamma}{\alpha} - 1$. Then, from (18) we have

$$\langle R_z(t)\rangle = -\frac{N}{2} + (N + \frac{\gamma}{\alpha})\{1 + e^{(t-\tilde{t}_0)/\tau_N}\}^{-1}, \quad \tau_N = \frac{N}{\alpha}(N + \frac{\gamma}{\alpha})^{-2},$$

In this particular case the radiation intensity (14) is

$$I(t) = \frac{\hbar\Omega\alpha}{4N}(N + \frac{\gamma}{\alpha})^3 \operatorname{sech}^2(\frac{t - \tilde{t}_0}{2\tau_N}) . \quad (19)$$

The parameter $\tilde{t}_0$ defines the position of the radiation-intensity maximum. For a special case of $\langle R_z\rangle_0 \equiv \langle R_z(0)\rangle = N/2$ we have $\tilde{t}_0 = \tau_N \ln(N\alpha/\gamma)$.

The right-hand side of (19) contains the term proportional to $N^2\alpha$. Such a term corresponds to the superradiation contribution to the radiation process.

In a more general case of $\beta^{-1} \geq 0$ instead of (17) from (16) we get

$$\frac{d}{dt}\langle R_z\rangle = -\frac{\alpha}{N}\langle R_z\rangle^3 + \{\frac{\alpha(M+1)}{N} + \frac{\eta}{N} + \frac{\gamma + 2n}{N}\}\langle R_z\rangle^2 +$$

$$+ \{\frac{\alpha N}{4} + \frac{\gamma}{2} + \frac{n}{N} - \frac{(\gamma + 2n)(M+1)}{N}\}\langle R_z\rangle - (\frac{n}{2} + \gamma\frac{M+1}{2} + \alpha N\frac{M+1}{2} + \frac{\eta N}{4}) , \quad (20)$$

where $\eta \equiv \sum_{\vec{\nu}} n^-_{\vec{\nu}}\{\Gamma(\vec{\nu} - \vec{\nu}_0) - N^{-1}\}$. Let X_1, X_2, X_3 be roofs of the polynomial in the right-hand side of (20). Then, its solution can be represented in the following form

$$e^{-\alpha(t-\tilde{t}_0)/N} = |\langle R_z\rangle - X_1|^{A_1} \cdot |\langle R_z\rangle - X_2|^{A_2} \cdot |\langle R_z\rangle - X_3|^{A_3} . \quad (21)$$

Here $A_1^{-1} = (X_1 - X_2)(X_1 - X_3)$, $A_2^{-1} = (X_2 - X_1)(X_2 - X_3)$, $A_3^{-1} = (X_3 - X_1)(X_3 - X_2)$.

This solution (21) describes the radiation or absorption processes in dependence on the initial state of the system.

For example, if $M = \frac{N}{2} - 1$, then from (20) we have

$$\frac{d}{dt}\langle R_z\rangle = -\frac{\alpha}{N}(\langle R_z\rangle - \frac{N}{2})(\langle R_z\rangle - X_+)(\langle R_z\rangle - X_-) , \quad (22)$$

where

$$X_\pm = \frac{\eta + \gamma + 2n}{2\alpha} \pm \sqrt{(\frac{\eta + \gamma + 2n}{2\alpha})^2 + \frac{N^2}{4} + \frac{\eta N + \gamma N + 2n}{2\alpha}} .$$

It should be noted that $X_+ > \frac{N}{2} > 0 > X_- > -\frac{N}{2}$. On the other hand, $-\frac{N}{2} < \langle R_z\rangle < M = \frac{N}{2} - 1$. So, if $\langle R_z\rangle_0 > X_-$ then $\frac{d}{dt}\langle R_z\rangle < 0$ and equation (22) describes the radiation process with $\langle R_z\rangle \xrightarrow[t\to\infty]{} X_-$. Its intensity $I(t) > 0$. If $\langle R_z\rangle_0 < X_-$, then $\frac{d}{dt}\langle R_z\rangle > 0$ and there is an absorption process in the system under consideration. In this case $I(t) < 0$ and $\langle R_z\rangle \to X_-$ when $t \to \infty$.

Let is now consider the case of $\mu = 1$. Then from (11) one can obtain for $\mathcal{O} = R_z$

$$\frac{d}{dt}\langle R_z\rangle = -N^{-1}\sum_{\vec{\nu}}\langle (S_{\vec{\nu}} + \frac{N}{2})\{bb^+(\gamma^+_{\vec{\nu}} - \gamma^-_{\vec{\nu}}) + n^+_{\vec{\nu}} - n^-_{\vec{\nu}}\}\rangle -$$

$$- N^{-1}\sum_{\vec{\nu}}\langle R_z bb^+\rangle(\gamma^+_{\vec{\nu}} + \gamma^-_{\vec{\nu}} + 2n^+_{\vec{\nu}} + 2n^-_{\vec{\nu}}) + N^{-1}\sum_{\vec{\nu}}\langle R_z\rangle(n^+_{\vec{\nu}} + n^-_{\vec{\nu}}) . \quad (23)$$

This equation differs from (15) by the so-called counter-rotation terms.

Let us suppose that the field mode $\vec{k}_b$ is so strong in the active volume V_c that $\langle b^+b\rangle =$ const. Then, $\langle S_{\vec{\nu}} bb^+\rangle \simeq \langle S_{\vec{\nu}}\rangle\langle bb^+\rangle$, $\langle R_z bb^+\rangle \simeq \langle R_z\rangle\langle bb^+\rangle$. Now, from (23) we get

$$\frac{d}{dt}\langle R_z\rangle = -\sum_{\vec{\nu}} G_{\vec{\nu}}(\langle S_{\vec{\nu}}\rangle + \frac{N}{2}) - Q\langle R_z\rangle , \quad (24)$$

where

$$G_{\vec{\nu}} \equiv N^{-1}\{\langle bb^+\rangle(\gamma^-_{\vec{\nu}} - \gamma^+_{\vec{\nu}}) + n^-_{\vec{\nu}} - n^+_{\vec{\nu}}\},$$

$$Q \equiv N^{-1}\sum_{\vec{\nu}}\{\langle bb^+\rangle(\gamma^-_{\vec{\nu}} + \gamma^+_{\vec{\nu}} + 2n^-_{\vec{\nu}} + 2n^+_{\vec{\nu}}) - (n^-_{\vec{\nu}} + n^+_{\vec{\nu}})\} .$$

As it is to be expected, equation (24) coincides with the equation for the intensity of a single-photon process to an approximation of the coefficients /3,4/.

Let the initial state of the system be the so-called Bloch state /10/. Then $\langle S_{\vec{\nu}}\rangle \simeq \{\Gamma(\vec{\nu} - \vec{\nu}_0) - N^{-1}\}(N^2/4 - \langle R_z\rangle^2)$

and (24) transforms into the following equation

$$\frac{d}{dt}\langle R_z\rangle = -\bar{G}(N^2/4 - \langle R_z\rangle^2) - Q\langle R_z\rangle - NU/2\,, \tag{25}$$

where $\bar{G} = \sum_{\vec{\nu}}\{\Gamma(\vec{\nu}-\vec{\nu}_0) - N^{-1}\}G_{\vec{\nu}}\,,\ U = \sum_{\vec{\nu}} G_{\vec{\nu}}\,.$

Its solution is

$$\left|\frac{\langle R_z\rangle - Y_+}{\langle R_z\rangle - Y_-}\right| = e^{\xi t}\cdot\text{const}, \quad \xi \equiv (N^2\bar{G}^2 + 2N\bar{G}U + Q^2)^{1/2}, \quad Y_{\pm} = \frac{Q \pm \xi}{2\bar{G}}\,.$$

Because $\bar{G} < U < Q$, $Y_+ > \frac{N}{2} > 0 > Y_- > -\frac{N}{2}$. For $\langle R_z\rangle_0 > Y_-$ solution of (25) is

$$\langle R_z\rangle = Y_- + \frac{Y_+ - Y_-}{1 + e^{\xi(t-\bar{t}_0)}}\,.$$

The corresponding intensity is

$$I(t) = \frac{\hbar\Omega}{4}(Y_+ - Y_-)\,\xi\,\text{sech}^2\{\tfrac{\xi}{2}(t-\bar{t}_0)\} > 0\,.$$

This intensity corresponds to the radiation process and contains the superradiation term $\sim N^2\bar{G}$. For $\langle R_z\rangle_0 < Y_-$ we have

$$\langle R_z\rangle = Y_- + \frac{Y_+ - Y_-}{1 - e^{\xi(t+\bar{t}_0)}}$$

and the formal expression for the intensity is

$$I(t) = -\frac{\hbar\Omega}{4}(Y_+ - Y_-)\,\xi\,\text{cosech}^2\{\tfrac{\xi}{2}(t+\bar{t}_0)\}\,.$$

It corresponds to the absorption process.

4. HIERARCHY FOR A COMBINATION SCATTERING PROCESS

In this section we shall consider the case described by Hamiltonian (4). Let us suppose that the initial conditions have a form (7). Then, the expounded above methos of partial elimination of the boson variables can be used again. So, the hierarchy of the kinetic equation can be obtained in the same manner as in section 2. Instead of (11) we have now

$$\frac{d}{dt}\langle \mathcal{O}\rangle + \frac{i}{\hbar}\langle[\mathcal{O}, H_{\bar{E}}]\rangle =$$

$$= \frac{1}{2N}\sum_{\vec{\nu}}\{\Gamma^+_{\vec{\nu}}\langle[R^-_{\vec{\nu}}b^+, \mathcal{O}]R^+_{\vec{\nu}}b\rangle + N^+_{\vec{\nu}}\langle[[R^-_{\vec{\nu}}b^+, \mathcal{O}], R^+_{\vec{\nu}}b]\rangle + \text{h.c.}\}\,. \tag{26}$$

Here all the notation is the same as in section 2 with the substitution of $\phi(\vec{k} - \vec{k}_b + \vec{\nu})$ for $\phi(\vec{k} + \vec{k}_b + \vec{\nu})$ and of $(-\omega_b)$ for ω_b. By analogy with (12) one can get the following equation

$$\frac{d}{dt}\langle R_z\rangle = -\frac{d}{dt}\langle b^+b\rangle = N^{-1}\sum_{\vec{\nu}}\{\gamma^+_{\vec{\nu}}\langle R^-_{\vec{\nu}}R^+_{\vec{\nu}}b^+b\rangle - n^+_{\vec{\nu}}\langle 2R_z bb^+ + R^-_{\vec{\nu}}R^+_{\vec{\nu}}\rangle\}\,. \tag{27}$$

Let us suppose that at the initial time moment there is only the pumping mode of field with the wave-vector $\vec{k}_b$ and $\beta^{-1} = 0$. Then instead of (27) we get

$$\frac{d}{dt}\langle R_z\rangle = -\frac{d}{dt}\langle b^+b\rangle = N^{-1}\sum_{\vec{\nu}}\gamma^+_{\vec{\nu}}\langle R^-_{\vec{\nu}}R^+_{\vec{\nu}}b^+b\rangle\,. \tag{28}$$

This equation has an integral of motion $\langle b^+(t)\,b(t)\rangle + \langle R_z(t)\rangle = \text{const} \equiv M$. In the case of a weak emitter-field correlation one can put

$$\langle R^-_{\vec{\nu}}R^+_{\vec{\nu}}b^+b\rangle \cong \langle R^-_{\vec{\nu}}R^+_{\vec{\nu}}\rangle\langle b^+b\rangle = \langle R^-_{\vec{\nu}}R^+_{\vec{\nu}}\rangle(M - \langle R_z\rangle)\,.$$

Let us suppose that at the initial moment all emitters are in the Bloch state and that the life-time of oscillators T is much longer than the duration of the process. Then

$$\langle R^-_{\vec{\nu}}R^+_{\vec{\nu}}\rangle \cong N/2 - \langle R_z\rangle + \{\Gamma(\vec{\nu}-\vec{\nu}_0) - N^{-1}\}(N^2/4 - \langle R_z\rangle^2)\,.$$

Here $\Gamma(\cdot)$ is the same as in section 3 and vector ν_0 characterises initial state of emitters. Now from (28) we obtain

$$\frac{d}{dt}\langle R_z\rangle = \frac{a}{N}(\langle R_z\rangle - M)(\langle R_z\rangle - \frac{N}{2})(\langle R_z\rangle + \frac{N}{2} + \frac{\gamma}{a}), \tag{29}$$

where $\gamma = \sum_{\vec{\nu}}\gamma^+_{\vec{\nu}}$, $a \equiv \sum_{\vec{\nu}}\gamma^+_{\vec{\nu}}\{\Gamma(\vec{\nu}-\vec{\nu}_0) - N^{-1}\}$ (compare with (17)).

The solution of equation (29) can be represented in the form which coincides with (18)

$$e^{a(t-\bar{t}_0)/N} = |\langle R_z\rangle - M|^{A_1}\cdot|\langle R_z\rangle - \frac{N}{2}|^{B_1}\cdot|\langle R_z\rangle + \frac{N}{2} + \frac{\gamma}{a}|^{C_1}, \tag{30}$$

where $A_1^{-1} \equiv (M - \frac{N}{2})(M + \frac{N}{2} + \frac{\gamma}{a})$, $B_1^{-1} \equiv (\frac{N}{2} - M)(N + \frac{\gamma}{a})$, $C_1^{-1} \equiv (M + \frac{N}{2} + \frac{\gamma}{a})(N + \frac{\gamma}{a})$.

Let at the initial point $\langle R_z\rangle_0 = -N/2$, $\langle b^+b\rangle = N$. Then $M = N/2$, and from (29) one can obtain

$$\langle R_z\rangle = \frac{N}{2} - \frac{N+\gamma/a}{1+\exp\{(t-\bar{t}_0)/r_N\}},$$

$$r_N^{-1} = aN^{-1}(N+\gamma/a)^2, \quad \bar{t}_0 = r_N \ln\frac{Na}{\gamma}.$$

So the rate of the pumping alteration is

$$\frac{d}{dt}\langle b^+b\rangle = -\frac{a}{4N}(N+\frac{\gamma}{a})^3 \operatorname{sech}^2(\frac{t-\bar{t}_0}{2r_N}).$$

This result coincides with (19).

Let us now analyse another case when $M \gg \frac{N}{2}$. It is obvious that $\langle R_z\rangle \to \frac{N}{2}$ as $t\to\infty$ and therefore $\langle b^+b\rangle \to M - \frac{N}{2} \cong M$. So $\langle b^+b\rangle \cong M$ and in (30) $A_1 \cong 0$, $B_1 \cong -C_1 \cong -\{M(N+\gamma/a)\}^{-1}$. Then, it follows from (30) that

$$\langle R_z\rangle = N/2 - \frac{N+\gamma/a}{1+\exp\{(t-\bar{t}_0)/r_N\}},$$

$$\frac{d}{dt}\langle b^+b\rangle = -\frac{a}{4N}(N+\frac{\gamma}{a})^2 M \operatorname{sech}^2(\frac{t-\bar{t}_0}{2r_N}),$$

where $r_N^{-1} = a(N+\gamma/a)M/N \cong aM \cong a\langle b^+b\rangle$. So, if the pumping is strong enough the duration of the process r_N is inversely proportional to the pumping intensity.

Now, we shall consider the case of $\beta^{-1} > 0$ and of a strong enough, so that $\langle b^+b\rangle \cong$ const. Then, from (26) we obtain

$$\frac{d}{dt}\langle R_z\rangle = -F_1\langle R_z\rangle^2 - F_2\langle R_z\rangle + (\frac{N^2F_1}{4} + \frac{NF_3}{2}), \tag{31}$$

where

$$F_1 \equiv N^{-1}\sum_{\vec{\nu}} \{\Gamma(\vec{\nu}-\vec{\nu}_0) - N^{-1}\}(\gamma^+_{\vec{\nu}}\langle b^+b\rangle - n^+_{\vec{\nu}}),$$

$$F_2 \equiv N^{-1}\sum_{\vec{\nu}} \{(\gamma^+_{\vec{\nu}} + 2n^+_{\vec{\nu}})\langle b^+b\rangle + n^+_{\vec{\nu}}\},$$

$$F_3 \equiv N^{-1}\sum_{\vec{\nu}} \{\gamma^+_{\vec{\nu}}\langle b^+b\rangle - n^+_{\vec{\nu}}\}.$$

The solution of (31) is

$$\left|\frac{\langle R_z\rangle - Y_-}{\langle R_z\rangle - Y_+}\right| = e^{t/r_N}\cdot \text{const}, \tag{32}$$

where $r_N^{-1} = \sqrt{N^2F_1^2 + 2NF_1F_3 + F_2^2}$, $Y_{\mp} = \dfrac{-F_2 \mp r_N^{-1}}{2F_1}$. Let us consider the following possibilities:

1. The pumping is strong, so that $F_1 > 0$. So $Y_- < -\frac{N}{2} < Y_+ < \frac{N}{2}$. If $\langle R_z\rangle_0 > Y_+$, then one can obtain from (32)

$$\langle R_z\rangle = Y_+ + \frac{Y_- - Y_+}{1-\exp\{(t+\bar{t}_0)/r_N\}},$$

and

$$\frac{d}{dt}\langle b^+b\rangle = -\frac{d}{dt}\langle R_z\rangle =$$
$$= \frac{1}{4F_1}(N^2F_1^2 + 2NF_1F_3 + F_2^2)\operatorname{cosech}^2(\frac{t+\bar{t}_0}{2r_N}). \tag{33}$$

This expression (33) describes the pumping amplification process. If $\langle R_z\rangle_0 < Y_+$, then

$$\langle R_z\rangle = Y_+ + \frac{Y_- - Y_+}{1+\exp\{(t-\bar{t}_0)/r_N\}},$$

and

$$\frac{d}{dt}\langle b^+b\rangle = -\frac{d}{dt}\langle R_z\rangle =$$
$$= -\frac{1}{4F_1}(N^2F_1^2 + 2NF_1F_3 + F_2^2)\operatorname{sech}^2\{(t-\bar{t}_0)/2r_N\}. \tag{34}$$

This equation corresponds to the weakening of pumping and to the Stokes-component radiation.

2. The temperature β^{-1} is so high that n^+_{ν} has a large value and $F_1 < 0$. Then $Y_- > \frac{N}{2} > Y_+ > -\frac{N}{2}$. For $\langle R_z\rangle_0 > Y_+$ there is the pumping amplification in the system:

$$\langle R_z\rangle = Y_+ + \frac{Y_- - Y_+}{1+\exp\{(t-\bar{t}_0)/r_N\}},$$

$$\frac{d}{dt}\langle b^+ b\rangle = -\frac{d}{dt}\langle R_z\rangle =$$

$$= \frac{1}{4|F_1|}(N^2F_1^2 + 2NF_1F_3 + F_2^2)\,\mathrm{sech}^2\{(t-\bar{t}_0)/2\tau_N\}. \quad (35)$$

For $\langle R_z\rangle_0 < Y_+$ there is the Stokes-component radiation and the weakening of pumping

$$\langle R_z\rangle = Y_+ + \frac{Y_- - Y_+}{1-\exp\{(t+\bar{t}_0)/\tau_N\}},$$

$$\frac{d}{dt}\langle b^+ b\rangle = -\frac{d}{dt}\langle R_z\rangle =$$

$$= -\frac{1}{4|F_1|}(N^2F_1^2 + 2NF_1F_3 + F_2^2)\,\mathrm{cosech}^2\{(t+\bar{t}_0)/2\tau_N\}. \quad (36)$$

It should be emphasised that equations (26) (or (11) in the case of cascade two-photon process) permit one to obtain some other information including the wave propagation in matter.

Here for simplicity we shall consider the case of zero temperature $\beta^{-1} = 0$. It can be easily shown that

$$\sum_{\vec{\nu}} |\phi(\vec{k}-\vec{k}_b+\vec{\nu})|^2 = 1, \quad \sum_{\vec{\nu}} |\phi(\vec{k}-\vec{k}_b+\vec{\nu})|^2\, |\phi(\vec{\nu}-\vec{\nu}_0)|^2 = \Gamma(\vec{k}-\vec{k}_b+\vec{\nu}_0).$$

Then

$$\gamma = \frac{4TV}{(2\pi)^3 N}\int d\vec{k}\,\frac{|g_{\vec{k}}|^2}{1+4T^2(\omega_k-\omega_b+\Omega)^2},$$

$$\alpha = \frac{4TV}{(2\pi)^3 N}\int d\vec{k}\,\frac{|g_{\vec{k}}\phi(\vec{k}-\vec{k}_b+\vec{\nu}_0)|^2}{1+4T^2(\omega_k-\omega_b+\Omega)^2} - \frac{\gamma}{N}.$$

Therefore from (29) we obtain

$$\frac{I(t)}{\hbar\Omega} = -\frac{d}{dt}\langle b^+ b\rangle = \frac{d}{dt}\langle R_z\rangle = \int d\vec{k}\, I_{\vec{k}}(t)\frac{1}{\hbar\Omega} =$$

$$= \frac{4TV}{(2\pi)^3 N}\int d\vec{k}\,\frac{f_{\vec{k}}}{1+4T^2(\omega_k-\omega_b+\Omega)^2}\cdot\frac{1}{N}\cdot(\langle R_z\rangle - M)(\langle R_z\rangle - \frac{N}{2}),$$

where

$$f_{\vec{k}} = |g_{\vec{k}}|^2\{1 + \{\Gamma(\vec{k}-\vec{k}_b+\vec{\nu}_0) - N^{-1}\}(\langle R_z\rangle + \frac{N}{2})\}. \quad (37)$$

The function $f_{\vec{k}}$ describes the time-dependent angular distribution. The first term in braces corresponds to the ordinary radiation process and the second to the collective one. At $N=1$ the collective contribution disappears. Let us consider that the emitters are rigidly fastened at lattice sites and $|g_{\vec{k}}|^2$ depends only on $|\vec{k}|$. For this case the ordinary radiation is isotropic, and the collective effects lead to the anisotropy: there is $\max I_{\vec{k}}(t)$ in the direction $\vec{k}\,||\,\vec{k}_b - \vec{\nu}_0$. This is the known condition of phase matching /11/.

Above we consider only the Stokes-component radiation process. For the case of anti-Stokes radiation (fig.2b) instead of (3) we have

$$H^{(as)}_{int} = \hbar N^{-1}\sum_j\sum_{\vec{k}}{}'\{g_{\vec{k}} e^{i(\vec{k}-\vec{k}_b)\vec{x}_j} a^+_{\vec{k}} b^+ r^+_j + g^*_{\vec{k}} e^{-i(\vec{k}-\vec{k}_b)\vec{x}_j} a_{\vec{k}} b\, r^-_j\}.$$

The corresponding hierarchy can also be obtained in the manner described above.

5. SUMMARY AND CONCLUSIONS

Exact hierarchies describing the two-photon kinetics of a two-level macroscopic system have been obtained in this paper. For this purpose the method of partial elimination of boson variables has been developed on the basis of Bogolubov's approach. This method consists in the following: First, we exclude boson variables for the modes (being initially on the equilibrium state) related to one step of the process. Thus, one can obtain an exact hierarchy of kinetic equations for the variables of the subsystem of emitters and separated residual mode related to the other step of the process (for instance to pumping). Under a further investigation of the obtained equations these residual variables can be eliminated at all by using various approximations or conservation laws.

The dynamics of the two-photon cascade process and of the process of the type of combination scattering has been studied within this method. Exact hierarchies of equations are found, and as a consequence, the Markovian equations. Different regimes of radiation and absorption are studied. Expressions for the intensity of radiation and absorption are found as well as for other characteristics. Angular distribution is obtained.

Our method can be applied for the investigation of other multi-photon processes. This will be a subject of further investigations.

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Боголюбов Н.Н./мл./, Фам Ле Киен, Шумовский А.С. E17-84-306
Динамика двухфотонных процессов
в двухуровневой системе

Развит метод частичного исключения бозонных переменных и на его основе получена точная иерархия для двухфотонных процессов в макроскопической двухуровневой системе. Исследована динамика каскадного процесса и процесса типа комбинационного рассеяния.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

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Bogolubov N.N.,Jr, Fam Le Kien, Shumovsky A.S. E17-84-306
Dynamics of Two-Photon Processes
in Two-Level System

A method of partial elimination of boson variables is developed and on its basis exact hierarchies for two-photon processes in a macroscopic two-level system are obtained. The dynamics of a cascade-type and of combination scattering processes is examined.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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