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Cooperative Spontaneous Lasing and Possible Quantum Retardation Effects

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Abstract

The collective decay effects between the dipole-active three-level subsystems in the nonlinear interaction with dipole-forbidden transitions, like $2S - 1S$ of hydrogen-like radiators, are proposed, taking into consideration the cooperative exchanges between two species of atoms through the vacuum field in the scattering and the two-photon resonance processes. One of them corresponds to the situation when the total energy of the emitted two photons by the three-level radiator in the cascade configuration enters into the two-photon resonance with another type of dipole-forbidden transitions of hydrogen-like (or helium-like) atoms. The similar situation appears in the cooperative scattering between two species of quantum emitters when the difference of the excited energies of the two dipole-active transitions of the three-level radiators is in the resonance with the dipole-forbidden transitions of the Hydrogen-like radiators. These effects are accompanied by the interference between single- and two-quantum collective transitions of the inverted radiators from the ensemble. The two-particle collective decay rate is defined in the description of the atomic correlation functions taking into consideration the phase retardation between them. The kinetic equations which describe the cooperative processes as the function of time and correlation are obtained. The behavior of the system of radiators at short and long time intervals in comparison with the retardation time between them is studied.

Keywords: 42.50.Fx Cooperative phenomena in quantum optical systems, 32.80.Qk Coherent control of atomic interactions with photons, 03.65.Ud Entanglement and quantum nonlocality, 03.65.Yz Decoherence, open systems, quantum statistical methods
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1. Introduction

The single-photon cooperative emission of the inverted system of radiators proposed by Dicke [1] opens the new possibilities of this phenomenon in the description of decay processes in the multilevel system [2] and multi-photon interaction of radiators with EMF (see, e.g., [3, 4]). The experimental possibilities [3, 4] of nonlinear cooperative interaction of radiators with vacuum field remain in the center of attention of many theoretical models proposed in the last time [5, 6].

For example, using the classical and quantum approaches in Refs. [7–10], it is given the quantitative description of two-color super-fluorescence, observed in [2]. In the recent experiment [11], the cooperative emission of excited atomic oxygen relatively the transition $3p^3P \rightarrow 3s^3S$ at wavelength $845nm$ as a result of two-photon photolysis of atmospheric O_2 followed by two-photon excitation of atomic oxygen by a laser pulse at $226nm$ is demonstrated.

Combining single- and two-photon processes, this chapter aims to investigate the cooperative emission of the inverted system of radiators taking into account the resonance between one- and two-photon cooperative transitions of two three-level atomic subsystems represented in **Figure 1**. In this approach, the two dipole-active species of radiators studied in Refs. [12, 13] are replaced with one three-level atomic subsystem Ξ (or V) inverted relative to the single-photon emission in the resonance with $2S$ - $1S$ dipole-forbidden transitions of hydrogen (or He)-like sub-ensemble. This new cooperative effect between two species of radiators occurs when two three-level emitters enter into two-quantum resonances with other emitters of the second ensemble inverted relative dipole-forbidden transition. Similar collectivization processes can amplify (or inhibit) the collective spontaneous emission rate of each atomic sub-ensemble. The sign of exchange integral between the two atoms from different sub-ensembles depends on the retardation time and distance between them. This problem is connected to the possibilities of amplifying of entangled quanta and established the coherence between photon pairs. For this, the cooperative interaction of three-radiator subsystems is proposed in which one of them is inverted relative to the dipole-forbidden transitions, but another inverted dipole-active three-level system ignites this transition.

Taking into consideration the elementary acts of two-photon resonance between radiators, we have demonstrated the increasing of two-photon emission rate in one of the radiator subsystem comparison with traditional two-photon super-fluorescence [5]. The mutual influence of two- and single-photon super-fluorescent processes on the two-photon cooperative emission of the inverted subsystem relatively dipole-forbidden transition depends on the position of atoms in the exchange potential. Two possibilities of two- and three-particle exchanges through the vacuum field are represented in **Figure 1A–C**, taking into consideration the two-
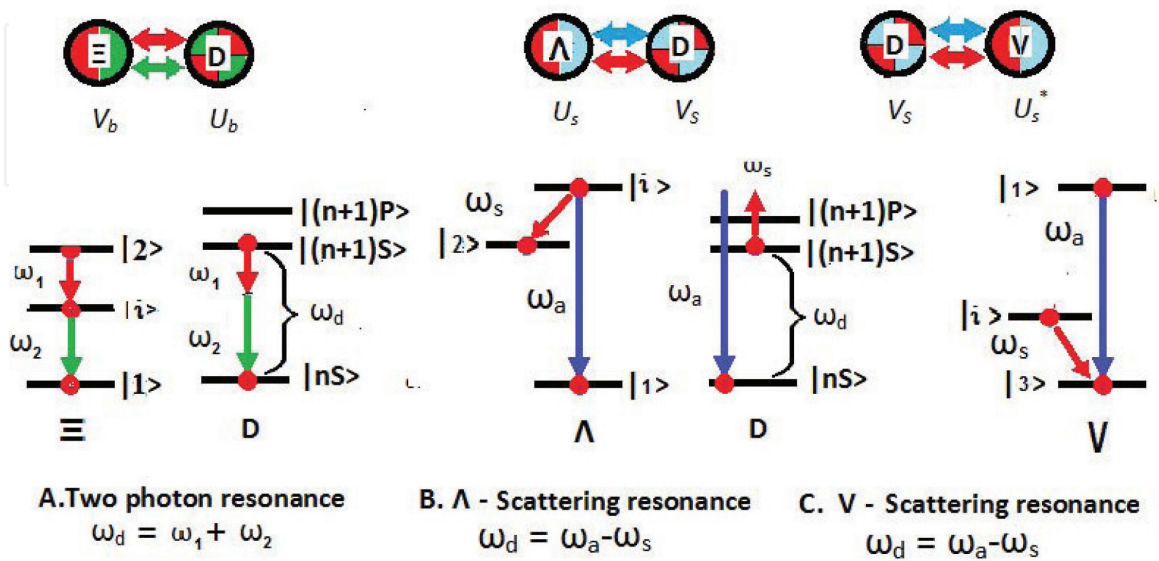
Figure 1.
The resonances between the two-photon transitions of D atomic subsystem and the three-level dipole-active systems in Ξ (A), V (B), and Λ (C) configurations. The three-level atoms are situated at relative distances $r_{d\xi}$, $r_{d\lambda}$, and r_{rv}. The exchange energies between the D subsystem in the two-photon resonance $\omega_0 = \omega_1 + \omega_2$ with the Ξ subsystem (A) and the scattering resonance $2\omega_0 = \omega_a - \omega_s$ with V subsystem (B) are given by the expressions (14) and (17).

photon resonance and scattering processes between the dipole-forbidden subsystem D and dipole-active subsystems of Ξ, Λ, and V, respectively. Here, the product of two vacuum polarizations of the atom Ξ (or V, Λ) comes into resonance with the polarization of the dipole-forbidden transitions of the D atom.

Using two small parameters in Section 2, we propose the projection operator method of elimination of the EMF operators from the generalized equation of atomic subsystems in single- and two-photon resonances. The possibilities of two-photon cooperative resonance between three-level radiators situated at a distance compared with the emission wavelength are demonstrated. Following this description the resonance interaction of a dipole-forbidden atom and three-level dipole-active radiator in the cascade configuration is described by the cooperative rate and the exchange integral (13). The similar expression (16) is obtained in the scattering process of three-level system in V or Λ— configurations with dipole-forbidden D subsystem represented in **Figure 1**. In Section 3 the spontaneous emission for the two radiators in the cascade or scattering resonances is given without the de-correlation of the atomic correlation functions between them.

2. Master equation of cooperative exchange between three-level radiators in two-quantum exchanges

Let us consider the interaction of three-level subsystems of radiators in V and Ξ configuration with D dipole-forbidden two-level ensemble through the vacuum of EMF. The Ξ three-level subsystem in cascade configuration, prepared in excited state $|2_\xi\rangle$, can pass into the Dicke super-radiance regime [1] relatively the dipole-active transitions $2_\xi \rightarrow \iota_\xi \rightarrow 1_\xi$ at frequencies ω_2 and ω_1 (**Figure 1A**). According to **Figure 1A**, the excited D atom relatively the dipole-forbidden transition $2_d \rightarrow 1_d$ passes in the ground state $|1_d\rangle$ simultaneously generating two quanta under the influence of cooperative decay of the Ξ three-level subsystem. Two-photon transition of the D-atom takes place through the virtual levels represented by the notations $|3_d\rangle$ with opposite parity relative to the ground $|1_d\rangle$ and excited $|2_d\rangle$ states, respectively. This case corresponds to the situation when the emission frequencies of the dipole-active Ξ radiators and D dipole-forbidden radiators satisfy the resonance condition $\omega_1 + \omega_2 = 2\omega_0$. Here ω_1 and ω_2 are the transition frequencies of the Ξ dipole-active radiators in Ξ, and $\hbar\omega_d = 2\hbar\omega_0$ is the energy distance between the ground $|nS\rangle$ and excited $|(n+1)S\rangle$ states of the dipole-forbidden transitions of D radiator (see **Figure 1A**).

The similar cooperative emissions can be observed in the two-quantum resonance interactions between the V (or Λ) three-level radiator in two quanta scattering interactions and the dipole-forbidden transitions of D atoms through the vacuum field (see **Figure 1B,C**). In this situation, we consider that the dipole-active transitions of the three-level radiator in the V (or Λ) configuration satisfy the scattering condition $\omega_a - \omega_s = \omega_d$ in interaction with the D subsystem. As it is represented in **Figure 1B**, the cone of the transition energies of the V or Λ dipole-active three-level atoms must be larger than the dipole-forbidden transition $|(n+1)S\rangle - |nS\rangle$ of atoms D, so that two-photon resonance between the two dipole-active transitions of V atom enters in the exact scattering resonance, $\omega_a - \omega_s = \omega_d$, with D atom. This nonlinear transition increases with the decreasing of the detuning from resonance with virtual $|3_d\rangle$ states of the D two-level system.

The Hamiltonian of the system consists of the free and interaction parts $H = \hat{H}_0 + \hat{H}_I$. Here the free part of this Hamiltonian is represented through the atomic and field operators:

$$\hat{H}_0 = \sum_k \hbar\omega_k \hat{a}_k^\dagger \hat{a}_k + \hbar \sum_{m=1}^{N} \omega_d \hat{D}_{zm} - \sum_{\alpha=s,}^{2} \sum_{al=1}^{N_\lambda} \hbar\omega_\alpha \hat{\Lambda}_{al}^\alpha$$

$$+ \sum_{\alpha=s,}^{2} \sum_{al=1}^{N_v} \hbar\omega_\alpha \hat{V}_{al}^\alpha + \sum_{\alpha=1,}^{N_\xi} \sum_{2j=1} \hbar(-1)^\alpha \omega_\alpha \hat{\Xi}_{\alpha j}^\alpha, \tag{1}$$

where N, N_ξ, N_λ, and N_v are the number of atoms in the D, Ξ, Λ, and V subsystems, respectively; the energies of first and second levels of the Ξ, Λ, and V three-level subsystems are measured from the third intermediate state $|\iota\rangle$. The operators $\Xi_{1,j}^1$, $\Xi_{\iota,j}^\iota$, and $\Xi_{2,j}^2$ describe the population of the ground, intermediary, and excited states of the Ξ atom. The population operators of two excited and ground states $\hat{V}_{2,j}^2$, $\hat{V}_{1,j}^1$, and $\hat{V}_{\iota,j}^\iota$ can be introduced for the three-level atom in V configuration too. The similar expressions for two ground and one excited state can be introduced for Λ three-level atomic configuration $\hat{\Lambda}_{2,j}^2$, $\hat{\Lambda}_{1,j}^1$, and $\hat{\Lambda}_{\iota,j}^\iota$, respectively. The D atoms are considered as a two-level system, the state energy positions of which are measured from the middle point between the excited and ground states, respectively, $D_{z,j} = \left(D_{2,j}^2 - D_{1,j}^1\right)/2$. The first term of the Hamiltonian describes the free energy of EMF, the $k \equiv$ k, λ modes of which is initially considered in the vacuum state $|0_k\rangle$. Here $\hat{a}_k$ and $\hat{a}_k^\dagger$ are annihilation and creation operators of EMF photons with wave vector k, polarization ε_λ, and the frequency ω_k, which satisfy the commutation relation $\left[\hat{a}_k^\dagger, \hat{a}_{k'}^\dagger\right] = \delta_{k,k'}$.

Taking into consideration the conservation energy laws, $\hbar(\omega_1 + \omega_2) = 2\hbar\omega_0$ and $\hbar(\omega_a - \omega_s) = 2\hbar\omega_0$ (according to **Figure 1A–C**, respectively), we introduce the interaction Hamiltonian $\hat{H}_I = \hat{H}_{I1} + \hat{H}_{I2}$ of the Ξ, Λ, V, and D subsystems with free EMF. Here $\hat{H}_{I1}$ describes the single-photon interaction of three-level atoms in the Ξ, V, and Λ configurations with a vacuum of EMF:

$$\hat{H}_{I1} = -\sum_k \sum_{j=1}^{N_\xi} \left[(\mu_{1\iota}, g_k)\hat{\Xi}_{1j}^\iota + (\mu_{2\iota}, g_k)\hat{\Xi}_{\iota j}^2\right]\hat{a}_k \exp\left[i(\text{k}, \text{r}_j)\right]$$

$$-\sum_k \sum_{l=1}^{N_\lambda} \left[(\mu_{\iota 1}, g_k)\hat{\Lambda}_{1l}^\iota + (\mu_{\iota 2}, g_k)\hat{\Lambda}_{2l}^\iota\right]\hat{a}_k \exp\left[i(\text{k}, \text{r}_l)\right] \tag{2}$$

$$-\sum_k \sum_{l=1}^{N_v} \left[(\mu_{\iota 1}, g_k)\hat{V}_{\iota l}^1 + (\mu_{\iota 2}, g_k)\hat{V}_{\iota l}^2\right]\hat{a}_k \exp\left[i(\text{k}, \text{r}_l)\right] + H.c.,$$

where $\varepsilon_1 \hat{H}_{I1}^{\Xi_1-} \sim \hat{\Xi}_{1j}^\iota \hat{a}_k$ and $\varepsilon_1 \hat{H}_{I1}^{\Xi_2=} \sim \hat{\Xi}_{\iota j}^2 \hat{a}_k$ represent the two-photon cascade excitation of Ξ atom through the intermediary state $|\iota\rangle$; $\varepsilon_1 \hat{H}_{I1}^{S-} \sim \hat{\Lambda}_{2,j}^\iota \hat{a}_k$ (or $\varepsilon_1 \hat{H}_{I1}^{S-} \sim \hat{V}_{\iota,j}^2 \hat{a}_k$) and $\varepsilon_1 \hat{H}_{I1}^{A-} \sim \hat{\Lambda}_{1,j}^\iota \hat{a}_k$ ($\varepsilon_1 \hat{H}_{I1}^{A-} \sim \hat{V}_{\iota,j}^1 \hat{a}_k$) describe the excitation of Λ (or V) atom with the absorption of the photons with the energies $\hbar\omega_s$ and $\hbar\omega_a$, respectively. $\mu_{i,j}$ is dipole momentum transitions between the i and j states of the atoms. The second part of interaction Hamiltonian, $\hat{H}_{I2}$, describes the nonlinear interaction of the dipole-forbidden transition of D two-level system with vacuum field:

$$\hat{H}_{I2} = \sum_{k_1, k_2} \sum_{m=1}^{N} [q_s(\text{k}_1, \text{k}_2)\hat{D}_m^- \hat{a}_{k_2}^\dagger \hat{a}_{k_1}(1 - \delta_{k_1, k_2}) \exp\left[i(\text{k}_1 - \text{k}_2, \text{r}_m)\right]$$

$$- q_b(\text{k}_1, \text{k}_2)\hat{D}_m^+ \hat{a}_{k_2} \hat{a}_{k_1} \exp\left[i(\text{k}_1 + \text{k}_2, \text{r}_m)\right]] + H.c. \tag{3}$$

This interaction is expressed by two-photon emission terms $\varepsilon_2 \hat{H}_{12}^{b+} \sim \hat{D}_m^- \hat{a}_{k_2}^\dagger \hat{a}_{k_1}^\dagger$ and possible scattering of an emitted photon by the Ξ and V subsystems $\varepsilon_2 \breve{H}_{12} s\pm \sim \hat{D}_m^\mp \hat{a}_{k_2}^\dagger \hat{a}_{k_1}$. The excitation and lowering operators of V, Λ, and Ξ dipole-active three-level subsystems are described by the operators of $U(3)$ algebra, which satisfy the commutation relations $\left[\hat{U}_{\beta j}^\alpha, \hat{U}_{\alpha' l}^{\beta'} \right] = \delta_{l,j} \left\{ \hat{U}_{\alpha' j}^\alpha \delta_{\beta, \beta'} + \hat{U}_{\beta' j}^\beta \delta_{\alpha, \alpha'} \right\}$. Here the operator $\hat{U}_{\beta j}^\alpha$ is equivalent with V and Ξ operators, $\hat{V}_{\beta j}^\alpha$ and $\hat{\Xi}_{\beta j}^\alpha$, respectively. The inversion $\hat{D}_{lz}$ together with lowering and exciting $\hat{D}_j^\pm$ operators of D subsystem belongs to $SU(2)$ algebra: $\left[\hat{D}_{lz}, \hat{D}_j^\pm \right] = \pm \hat{D}_l^\pm \delta_{l,j}$ and $\left[\hat{D}_l^+, \hat{D}_m^+ \right] = 2\delta_{l,m} \hat{D}_{lz}$. In comparison with single-photon interaction of Ξ and V atoms with vacuum field $\left(\mu_{i,j}, g_k \right)$, the nonlinear interaction of D two-level subsystem with EMF in two-photon and scattering interaction is described by the interaction constants and second order:

$$q_b(k_1, k_2) = \frac{\left(d_{31}, g_{k_1} \right)\left(d_{32}, g_{k_2} \right)}{2\hbar(\omega_{32} + \omega_{k_1})} + \frac{\left(d_{31}, g_{k_2} \right)\left(d_{32}, g_{k_1} \right)}{2\hbar(\omega_{31} - \omega_{k_2})},$$

$$q_s(k_1, k_2) = \frac{\left(d_{31}, g_{k_1} \right)\left(d_{32}, g_{k_2} \right)}{\hbar(\omega_{32} - \omega_{k_1})} + \frac{\left(d_{31}, g_{k_2} \right)\left(d_{32}, g_{k_1} \right)}{\hbar(\omega_{31} + \omega_{k_1})},$$

where $g_k = \varepsilon_\lambda \sqrt{2\pi\hbar\omega_k / V}$ and $d_{i,j}$ is dipole momentum transitions between the levels of the D atom. In the definition of the interaction parts of the Hamiltonian (2) and (3), we introduced the fictive small parameters ε_1 and ε_2 which will help us to establish the contributions of the second and third orders in two-photon decay rates.

In this section the conditions for which the pure super-fluorescence of the small number of radiators [14, 15] in the subsystems Ξ, V, and D enters into interaction during the delay time of cooperative spontaneous emission of each subsystem are considered, so that inhomogeneous broadening of excited atomic states can be neglected, $\tau_i \ll T_{2,i}$. Here $\tau_i = \tau_0 / N_i$ is the collective time for which the polarization of the i subsystem becomes macroscopic; $T_{2,i}$ is the de-phasing time of the subsystem i, which includes the reciprocal inhomogeneous and Doppler-broadened line-width, $i \equiv \Xi$, V, and D (see, e.g., the papers [15, 16]). These conditions can be achieved using laser cooling method [17, 18] for three atomic ensembles represented in **Figure 1**A,B. Let us suppose that delay time of the super-radiant pulse is less than $T_{2,i}$; we will drop the terms connected with de-phasing time $T_{2,i}$ from the kinetic equations. In order to estimate the three-particle cooperative interaction, we will examine the situation in which one- and two-quantum interactions with the EMF bath are taken into account simultaneously. In this case it is necessary to eliminate from the density matrix equation the boson operators of EMF in nonlinear interaction with atomic subsystem. In comparison with the paper [12], here we will take into consideration the two-quantum effects connected with the influence of three-level atomic systems V and Ξ on the two-photon spontaneous emission of dipole-forbidden D subsystem. In this case instead of two dipole-active atoms, we can take into consideration only one three-level atom in two-photon resonance with dipole-forbidden system.

Let $\mathcal{P}$ be the projection operator for the complete density matrix $\breve{\rho}(t)$ on the vector basis of a free EMF subsystem $\rho_s(t) = \mathcal{P}\breve{\rho}(t)$ and $\breve{\rho}_b(t) = \overline{\mathcal{P}}\breve{\rho}(t)$, where $\breve{\rho}_s(t)$ and $\breve{\rho}_b(t)$ are slower and rapidly oscillating parts of the density matrix, respectively,

$\overline{\mathcal{P}} = 1 - \mathcal{P}$. It can be shown that $\mathcal{P}^2 = \mathcal{P}$ and $\overline{\mathcal{P}}\mathcal{P} = 0$. Recognizing that for $t = 0$ an electronic subsystem does not interact with the EMF, we define the projection operator $\mathcal{P} = \breve{\rho}_{ph}(0) \otimes Tr_{ph}\{\cdots\}$, where the trace is taking over the photon states and $\breve{\rho}_{ph}(0) = |0\rangle\langle 0|$ represents the density matrix of the vacuum of EMF. In this case one can represent the slow part of density matrix through the density matrix $\breve{W}(t) = Tr_{ph}\{\breve{\rho}(t)\}$ of the atomic subsystem $\breve{\rho}_s(t) = \breve{\rho}_{ph} \otimes \breve{W}(t)$, where $\breve{W}(0) = Tr_{ph}\breve{\rho}(0) = \breve{\rho}_r(0)$ is the density matrix of the prepared state of the atomic subsystem. The equations for the matrix $\breve{\rho}_s(t)$ and $\breve{\rho}_b(t)$ are

$$\frac{\partial \breve{\rho}_s(t)}{\partial t} = -i\mathcal{P}L_I(t)\{\breve{\rho}_s(t) + \breve{\rho}_b(t)\}, \tag{4}$$

$$\frac{\partial \breve{\rho}_b(t)}{\partial t} = -i\overline{\mathcal{P}}L_I(t)\{\breve{\rho}_s(t) + \breve{\rho}_b(t)\}, \tag{5}$$

where $\hat{L}_I(t) = \varepsilon_1[\breve{H}_{I1}(t),...]/\hbar + \varepsilon_2[\breve{H}_{I2}(t),...]/\hbar$ is the interaction part of Liouville operator. Following the known procedure of elimination of the rapidly oscillating part of the density matrix, we integrate Eq. (5) with respect to $\breve{\rho}_b(t)$ and substitute the resulting solution in Eq. (4). After this procedure we obtain the expression

$$\frac{\partial \breve{\rho}_s(t)}{\partial t} = -P \int_0^t d\tau L_I(t)U(t, t - \tau)L_I(t - \tau)\rho_s(t - \tau), \tag{6}$$

where the two-time evolution operator is represented by the T product $\breve{U}(t, t - \tau) = T \exp\left\{-i\overline{\mathcal{P}} \int_{t-\tau}^t d\tau_1 L_I(\tau)\right\}$. In comparison with well-known procedure of the decomposition on the small parameter ε of the right-hand site of expression (6), here we have two parameters ε_1 and ε_2. The quantum correlation between the single- and two-photon interactions of atoms through the vacuum of the EMF can be found in the third order of the expansion on the small parameter product $\varepsilon_1^2\varepsilon_2$ of the right-hand side of Eq. (6). Indeed considering the second and third order of the expansion on the small parameters ε_1 and ε_2, we represent the evolution operators $\breve{U}(t, t - \tau)$ and $\breve{\rho}_s(t - \tau)$ in the following approximate form $\breve{U}(t, t - \tau) \approx 1 - i\overline{\mathcal{P}} \int_{t-\tau}^t d\tau_1 L_i(\tau_1)$ and $\breve{\rho}_s(t - \tau) = \breve{\rho}_s(t) + \mathcal{P} \int_0^\tau d\tau_1 \hat{L}_i(t - \tau_1) \int_0^{t-\tau_1} d\tau_2 \hat{L}_i(t - \tau_1 - \tau_2)\breve{\rho}_s(t - \tau_1 - \tau_2)$. Upon substitution of this expression in Eq. (6), in the third order of small parameter λ, the equation for $\rho_s(t)$ becomes

$$\frac{\partial}{\partial t}\breve{\rho}_s(t) = -\mathcal{P} \int_0^t d\tau_1 \hat{L}_i(t)\left\{\hat{L}_i(t - \tau_1) - i \int_{t-\tau_1}^t d\tau_2 \hat{L}_i(\tau_2)\right\}\hat{L}_i(t - \tau_1)\breve{\rho}_s(t). \tag{7}$$

Representing the Liouville operator, $\hat{L}_I(t)$, through single-, $L_{I1}(t) = \varepsilon_1[\breve{H}_{I1}(t),...]/\hbar$, and two-photon, $\lambda L_{I2}(t) = \varepsilon_2[\breve{H}_{I2}(t),...]/\hbar$, interaction parts, we can observe that in the third order on the decomposition on interaction Hamiltonian, the main contribution to the right-hand site of Eq. (7) gives the terms proportional to the $\varepsilon_1^2\varepsilon_2$. Indeed, taking into consideration that the trace of an odd number of boson operator is zero, $Tr_{ph}\left\{\rho_0 \breve{a}_{k_1}^\dagger \breve{a}_{k_2}{}^\dagger \breve{a}_{k_3} \breve{a}_{k_4} \breve{a}_{k_5}\right\} = 0$, it is not difficult to observe

that the projection of the operator product $\varepsilon_2^2\varepsilon_1\mathcal{P}\breve{H}_{I1}\breve{H}_{I2}\breve{H}_{I2}$ takes the zero value too. In the third order of the small parameters ε_i, the contribution of Liouville operator $\hat{L}_{I1}$ and $\hat{L}_{I2}$ must be found from the terms like $\mathcal{P}\hat{L}_{I1}\hat{L}_{I2}\hat{L}_{I1}\mathring{\rho}_s(t)$, which corresponds to two-photon resonances between the single- and two-photon transitions in the three-level atomic systems described by the Hamiltonian part (2) and (3), respectively. It is not difficult to observe that second-order decomposition on the interaction Hamiltonian gives zero contributions in the correlations between the Ξ, V, and D subsystems. This follows from the zero value of the trace of the odd number of boson operators, $Tr_{ph}\left\{\breve{\rho}_0\breve{a}_{k_1}^\dagger\breve{a}_{k_2}\breve{a}_{k_3}\dagger\right\} = 0$, which corresponds to the projection of the operator product $\mathcal{P}\breve{H}_{I1}\breve{H}_{I2}\mathcal{P} = 0$.

Following this procedure of calculation of mean value of boson operators, it is observed that the two-photon resonance represented in **Figure 1A** can be described by the following diagrams:

$$\Delta\rho_3^b = i\lambda^3\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\Big\{\mathcal{P}\hat{L}_{I1}^{\Xi_1-}(t)\hat{\hat{L}}_{I1}^{\Xi_2-}(t-\tau_2)\hat{L}_{I2}^{b+}(t-\tau_1)\rho_s(t)$$

$$+\mathcal{P}\hat{L}_{I1}^{R-}(t)\hat{L}_{I1}^{S-}(t-\tau_2)\hat{L}_{I2}^{b+}(t-\tau_1)\rho_s(t)$$

$$+\mathcal{P}\hat{L}_{I1}^{\Xi_2-}(t)\hat{L}_{I2}^{b+}(t-\tau_2)\hat{L}_{I1}^{\Xi_1-}(t-\tau_1)\rho_s(t) \quad\quad (8)$$

$$+\mathcal{P}\hat{L}_{I1}^{\Xi_1-}(t)\hat{L}_{I2}^{b+}(t-\tau_2)\hat{L}_{I1}^{\Xi_2-}(t-\tau_1)\rho_s(t)$$

$$+\mathcal{P}\hat{L}_{I2}^{b+}(t)\hat{L}_{I1}^{\Xi_1-}(t-\tau_2)\hat{L}_{I1}^{\Xi_2-}(t-\tau_1)\rho_s(t)$$

$$+\mathcal{P}\hat{L}_{I2}^{b+}(t)\hat{L}_{I1}^{\Xi_2-}(t-\tau_2)\hat{L}_{I1}^{\Xi_1-}(t-\tau_1)\rho_s(t)\Big\} + H.c.$$

Here $L_{I1}^{\Xi_1-}(t) = \varepsilon_1\left[\breve{H}_{I1}^{\Xi_1-}(t),...\right]/\hbar$, $L_{I1}^{\Xi_2-}(t) = \varepsilon_1\left[\breve{H}_{I2}^{\Xi_2-}(t),...\right]/\hbar$, and $L_{I2}^{b-}(t) = \varepsilon_2\left[\breve{H}_{I2}^{b-}(t),...\right]/\hbar$ represent the Liouville operators of the interaction part of the Ξ and D atoms expressed through EMF annihilation and atomic exciting operators in the single- and two-quantum interactions.

The scattering resonance can be represented by the diagrams in which the conservation law $\omega_a - \omega_s = 2\omega_0$ must take place as represented in **Figure 1B**:

$$\Delta\rho_3^s = i\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\Big\{\mathcal{P}L_{I1}^{A-}(t)L_{I1}^{S+}(t-\tau_2)L_{I2}^{s+}(t-\tau_1)\rho_s(t)$$

$$+\mathcal{P}L_{I1}^{S+}(t)L_{I1}^{A-}(t-\tau_2)L_{I2}^{s+}(t-\tau_1)\rho_s(t)$$

$$+\mathcal{P}L_{I1}^{A-}(t)L_{I2}^{s+}(t-\tau_1)L_{I1}^{S+}(t-\tau_2)\rho_s(t) \quad\quad (9)$$

$$+\mathcal{P}L_{I1}^{S+}(t)L_{I2}^{s+}(t-\tau_1)L_{I1}^{A-}(t-\tau_2)\rho_s(t)$$

$$+\mathcal{P}L_{I2}^{s+}(t)L_{I1}^{A-}(t-\tau_1)L_{I1}^{S+}(t-\tau_2)\rho_s(t)$$

$$+\mathcal{P}L_{I2}^{s+}(t)L_{I1}^{S+}(t-\tau_1)L_{I1}^{A-}(t-\tau_2)\rho_s(t)\Big\} + H.c.,$$

where $L_{I2}^{s-}(t) = \varepsilon_2\left[\breve{H}_{I2}^{s-}(t),...\right]/\hbar$ is the Liouville parts for two-photon scarpering process of D atomic subsystem and $L_{I1}^{S-}(t) = \varepsilon_1\left[\breve{H}_{I1}^{S-}(t),...\right]/\hbar$ and

$L_{I1}^{A-}(t) = \varepsilon_1\left[\breve{H}_{I1}^{A-}(t), \ldots\right]/\hbar$ correspond to the single-photon transitions in Ξ atomic subsystem described by the Hamiltonian parts (3) and (2), respectively.

So that after the trace on the EMF variables, we obtain $Tr\left\{\hat{\rho}_{ph}\breve{a}_{k_1}\breve{a}_{k_3}\breve{a}_{k_2}\dagger\breve{a}_{k_4}\dagger\right\}$
$= \left[\delta_{k_1 k_2}\delta_{k_3, k_4} + \delta_{k_1, k_4}\delta_{k_3, k_2}\right]$, $Tr\left\{\hat{\rho}_{ph}\breve{a}_{k_1}\breve{a}_{k_2}\dagger\right\} = \delta_{k_1 k_2}$, and $Tr\left\{\rho_{ph}\breve{a}_{k_2}^\dagger\breve{a}_{k_4}\dagger\breve{a}_{k_1}\breve{a}_{k_3}\right\} = 0$.
We found the correlations between Ξ, V, and D atomic subsystem represented in **Figure 1**.

We found the correlations between Ξ, V, and D atomic subsystem represented in the **Figure 1**. Following projection technique procedures developed in Refs. [5, 13, 19], we find the terms of in the right-hand side of the master equation (7)–(9) for three species of radiators in interaction

$$\frac{d\breve{W}(t)}{dt} = \frac{d\breve{W}_0(t)}{dt} + \frac{d\breve{W}_{21b}(t)}{dt} + \frac{d\breve{W}_{21s}(t)}{dt}. \qquad (10)$$

First term describes the cooperative single- and two-photon effects in each subsystem, respectively. Second term describes the exchanges between the single-photon processes of Ξ three-level subsystem and the two-photon transitions of the D radiators as this is represented in **Figure 1A**. The third term describes the scattering effect of the two radiators represented in **Figure 1B**.

All parameters and collective exchange integrals between the three-level radiators in V configuration and dipole-forbidden two-level system D are defined in the literature [1–12]:

$$\frac{d\breve{W}_0(t)}{dt} = \frac{1}{2\tau_{\iota,1}}\sum_{l,j=1}^{N_\xi}\chi_1(j,l)\left[\breve{\Xi}_{\iota,j}^1, \breve{W}(t)\breve{\Xi}_{1,l}^\iota\right] + \frac{1}{2\tau_{\iota,2}}\sum_{l,j=1}^{N_\xi}\chi_2(j,l)\left[\breve{\Xi}_{2,j}^\iota, \breve{W}(t)\breve{\Xi}_{\iota,l}^2\right]$$

$$+ \frac{1}{2\tau_{\iota,a}}\sum_{l,j=1}^{N_v}\chi_a(j,l)\left[\breve{V}_{1,j}^\iota, \breve{W}(t)\breve{V}_{\iota,l}^1\right] + \frac{1}{2\tau_{\iota,s}}\sum_{l,j=1}^{N_v}\chi_s(j,l)\left[\breve{V}_{2,j}^\iota, \breve{W}(t)\breve{V}_{\iota,l}^2\right]$$

$$+ \frac{1}{2\tau_{\iota,s}}\sum_{l,j=1}^{N_\lambda}\chi_s(j,l)\left[\breve{\Lambda}_{\iota,j}^2, \breve{W}(t)\breve{\Lambda}_{2,l}^\iota\right] + \frac{1}{2\tau_{\iota,a}}\sum_{l,j=1}^{N_\lambda}\chi_a(j,l)\left[\breve{\Lambda}_{\iota,j}^2, \breve{W}(t)\breve{\Lambda}_{2,l}^\iota\right]$$

$$+ \frac{1}{2\tau_d}\sum_{l,j=1}^{N}\chi_d(j,l)\left[\breve{D}_j^-, \breve{W}(t)\breve{D}_l+\right] + H.c., $$

$$(11)$$

where $\tau_{\iota,\alpha} = 3\hbar c^3/\left(4\mu_{\alpha,\iota}^2\omega_\alpha^3\right)$ is the spontaneous emission time of the dipole-active transitions $|\alpha\rangle \to |\iota\rangle$ of three-level atom in Ξ and V configurations and $\tau_d = \pi 3^2\hbar^2 c^6/\left(4^2\omega_0^7 d_{23}^2 d_{31}^2 q_b^2(\omega_0,\omega_0)\right)$ is the two-photon spontaneous emission rate in the D atomic subsystem. This equation can be used for the description of interaction between the dipole-forbidden and dipole-active subsystems of radiators. For comparison of the real parts of the single- and two-photon exchange integrals, we can observe that the second decreases inversely proportional to the square distance r_{jl} between two D radiators: $\mathrm{Re}[\chi_\alpha(j,l)] = \sin\left(\omega_\alpha r_{j,l}/c\right)/\left[\omega_\alpha r_{j,l}/c\right]$ and $\mathrm{Re}[\chi_d(j,l)] \sim \sin^2(\omega_0 r_{j,l}/c)/\left[\omega_0 r_{j,l}/c\right]^2$.

Following the two-parameter approach projection technique proposed in Ref. [13], $H_I1 \sim \varepsilon_1$ and $H_I2 \sim \varepsilon_2$, we easily found the three-particle exchanges between the radiators represented in **Figure 1A** described by master equation

$$\frac{d\breve{W}_{21b}(t)}{dt} = -\frac{i}{4\tau_{12d}^{b}}\sum_{m=1}^{N}\sum_{l=1}^{N_\xi}\sum_{j=1}^{N_\xi}\{U_b(m,l)$$

$$\times\left\{\left[\breve{D}_m^-,\breve{W}(t)\breve{\Xi}_{i,l}^2\breve{\Xi}_{1,j}^l\right] + \left[\breve{D}_m^-,\breve{W}(t)\breve{\Xi}_{1,j}^l\breve{\Xi}_{i,l}^2\right]\right\}$$

$$+ U_b^*(j,l,m)\left[\breve{\Xi}_{i,l}^2,\left[\breve{\Xi}_{1,j}^l,\breve{D}_m-\breve{W}(t)\right]\right]$$

$$+ [\breve{\Xi}_{1,j}^l,[\breve{\Xi}_{i,l}^2,\breve{D}_m-\breve{W}(t)]]\} - \frac{i}{2\tau_{12d}^{b}}\sum_{m=1}^{N}\sum_{j=1}^{N_\xi}\sum_{l=1}^{N_\xi}V_b(j,m,l)$$

$$\times\left\{\left[\breve{D}_m^-\breve{W}(t)\breve{\Xi}_{il}^2,\breve{\Xi}_{1,j}^l\right] + \left[\breve{D}_m^-\breve{W}(t)\breve{\Xi}_{1,j}^l,\breve{\Xi}_{i,l}^2\right]\right\}$$

$$+ H.c. \tag{12}$$

Here for $\omega_s \simeq \omega_r$, we have found the following integrals:

$$\frac{1}{\tau_{12d}^{b}} = \left\{\frac{4}{3}\right\}^2\frac{\omega_s^3(\omega_r)^3\mu_{i2}\mu_{i1}d_{23}d_{31}}{2\hbar^2 c^6}\left[\frac{1}{\omega_{32}+\omega_2}+\frac{1}{\omega_{31}+\omega_1}\right],$$

$$V_b(j,m,l) \simeq \frac{c^2[\exp[-i\omega_2 r_{ml}/c]-1][\exp[i\omega_1 r_{jm}/c]-1]}{\omega_1\omega_2 r_{jm}r_{ml}}, \tag{13}$$

$$U_b(j,m,l) = \exp[-i\omega_1 r_{mj}/c]V_b(j,l,m).$$

Here $1/\tau_{12d}^{b}$ is the three-particle cooperative emission rate of two atoms from Ξ subsystems and one atom from D ensemble situated at the relatively small distance $r_{jl} \ll \lambda_{s(r)}$. $V_b(j,m,l)$ is the exchange integral which describes the influence of the m atom from D ensemble on the single-photon transitions of the j and l radiators from the Ξ subsystem. $U_b(j,m,l)$ is the inverse process of the cooperative action of j and l radiators from the Ξ ensemble on the two-photon transitions of m radiator from the D subsystem.
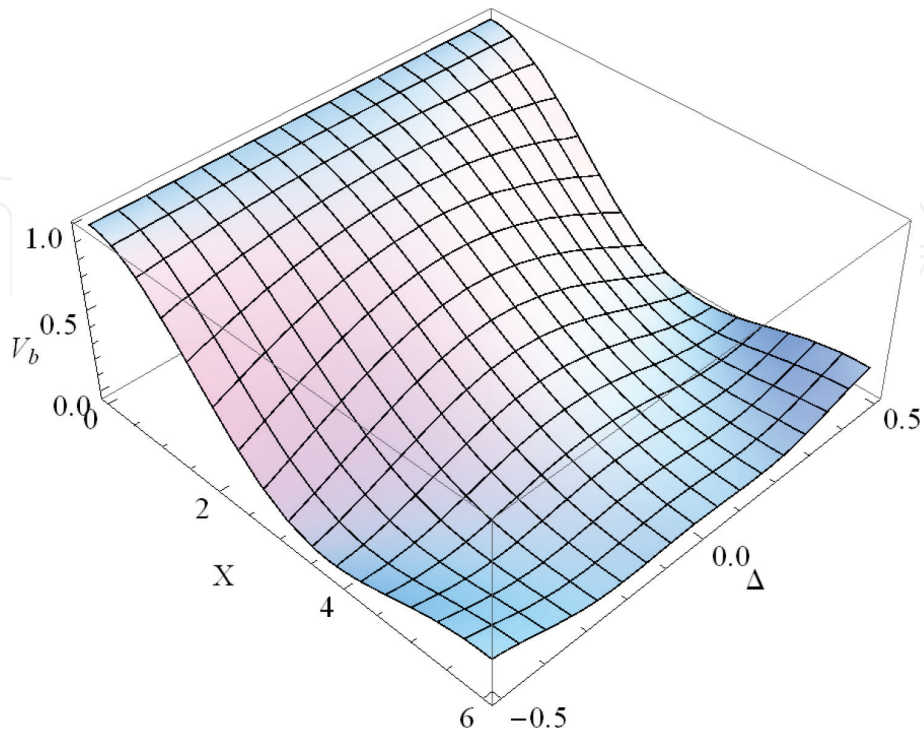
Figure 2.
The real part of exchange integral V_b, defined in expression (14), is plotted as a function of relative distance between radiators, $X = \omega_0 r/c$, and relative displacement, $\Delta = (\omega_1 - \omega_0)/\omega_0$.

For two atoms represented in **Figure 1A**, the simple exchange integral between these radiators can be obtained from expression (13):

$$V_b = \frac{\lambda_1 \lambda_2 [\exp(-2i\pi r/\lambda_2) - 1][\exp(2i\pi r/\lambda_1) - 1]}{(2\pi r)^2}, \qquad (14)$$

where λ_2 and λ_1 are the emission wavelengths in cascade transition of the dipole-active three radiators in Ξ configuration, situated at distance r. The real part of this function describes the three-particle decay rate of the system. The dependence of exchange integral (14) on the relative distance between the Ξ and D atoms (14), $X = \omega_0 r/c$ and the displacement, $\Delta = (\omega_1 - \omega_0)/\omega_0$ relatively the degenerate frequency ω_0, is plotted in **Figure 2**. As follows from this dependence, the exchange integral achieved the maximal radius, when $\omega_1 = \omega_2$, which corresponds to the situation $\Delta = 0$.

The part of master Equation (10) for resonance scattering interaction between the absorbed and emitted photons by the dipole-active Λ and V subsystems and D dipole-forbidden radiators can be obtained from the third-order expansion on the smallest parameter λ. In this situation, the scattering part of the master equation represented by the scheme 1 B becomes

$$
\begin{aligned}
\frac{d\breve{W}_{21s}(t)}{dt} = {} & \frac{i}{2\tau_{sad}^s} \sum_{m, j, l=1} \left\{ U_s(j,m,l)[\breve{V}_{i,j}^1, \breve{V}_{2,l}^l \breve{D}_m - \hat{W}(t)] \right. \\
& \left. + U_s^*(j,m,l)[\breve{V}_{2,l}^l, \breve{W}(t)\breve{V}_{i,j}^1 \breve{D}_m-] \right\} \\
& - \frac{i}{2\tau_{sad}^s} \sum_{m, j, l=1} V_s(j,m,l)\left[\breve{D}_m^-, \breve{V}_{2,l}^l \breve{W}(t)\breve{V}_{i,j}^1\right] \\
& + H.c.,
\end{aligned} \qquad (15)
$$
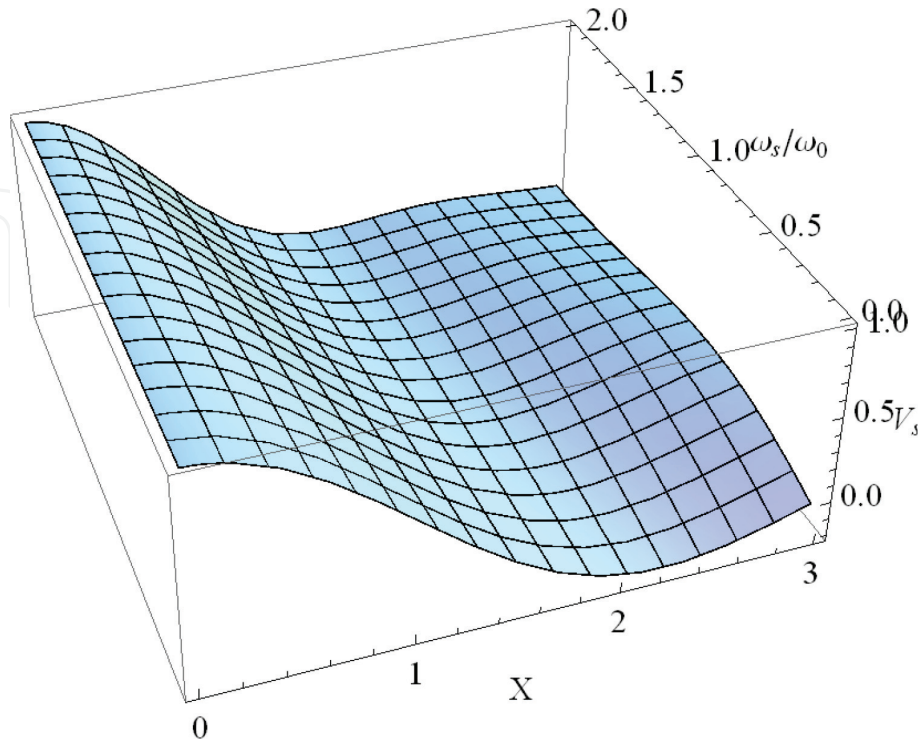
Figure 3.
The real part of the scattering exchange integrals V_s, defined in expressions (17), is plotted as a function of relative distance between radiators, $X = \omega_0 r/c$, and relative scattering frequency, ω_s/ω_0.

where

$$\frac{1}{\tau^s_{sad}} = \left(\frac{4}{3}\right)^2 \frac{\mu_{i2}\mu_{i1}d_{23}d_{31}\omega_s^3(\omega_a)^3}{c^6\hbar^2}\left[\frac{1}{\omega_{32}-\omega_s}+\frac{1}{\omega_{31}+\omega_s}\right],$$

$$V_s(j,m.l) = \frac{(\exp[-i\omega_s r_{ml}/c]-1)(\exp[i\omega_a r_{mj}/c]-1)}{\omega_s\omega_a(r_{ml}/c)(r_{mj}/c)}, \qquad (16)$$

$$U_s(j,m,l) = \exp[-i\omega_s r_{ml}/c]V_s(j,m.l).$$

First term in Eq. (15) describes the transition of D atom under the influence of the scattering process of emitted photons of the atoms from V subsystem. This process is described by exchange integral $V_s(j,m.l)$. The last two terms in master Eq. (15) describe the scattering process of emitted photons by the V atoms under the influence of D subsystem.

The similar expression is obtained for the interaction of Λ three-level radiator with D atom represented in **Figure 1C**. In this case we must replace the operators of V subsystem in expression (15) with corresponding transition operators of Λ system $\breve{V}^1_{i,j} \rightarrow \hat{\Lambda}^i_{1j}: \breve{V}^i_{2,l} \rightarrow \breve{\Lambda}^2_{i,l}$ and their Hermit conjugated operators.

For the two atoms, expression (16) was reduced to the simple representation

$$V_s = \frac{\lambda_s\lambda_a[1-\exp(-2i\pi r/\lambda_s)][1-\exp(2i\pi r/\lambda_a)]}{(2\pi r)^2}. \qquad (17)$$

Here the wavelength λ_s (λ_a) corresponds to the emitted photons at Stokes or anti-Stokes frequencies represented in **Figure 1**. The numerical representation of the real part of the exchange integral (17) as the function of the relive distance between the atoms $X = \omega_0 r/c$ and the relative Stokes frequency ω_s/ω_0 is plotted in the **Figure 3**. It is observing the nonsignificant dependence of this exchange integral on the frequency ω_s. The significant dependence on the detuning from resonance can be observed in the dependence of cooperative rate $1/\tau^s_{sad}$ represented in expressions (16).

In this section we obtained the correlations between dipole-active and dipole-forbidden subsystems of radiators, where the two-quantum exchange integral has the same magnitude as the two-photon quantum interaction between atoms of D subsystem. In the case of the big number of radiators in each subsystem, the correlated terms, expressions (12) and (15), give the cubic contribution in the cooperative diagrams of the kinetic equation $\varepsilon_1^2\varepsilon_2 NN_\xi^2$. When $N = N_\xi$ these terms can archived the value proportional to the Dicke super-radiance [1] even for the same small parameters of each subsystem $\varepsilon_1 = \varepsilon_2$. In this case the number of atoms in each subsystem must achieve the value for which the third order has the same magnitude as the second order $\varepsilon^2 N^2 \sim \varepsilon^3 N^3$. In conclusion we observe that the decomposition on the small parameter ε can be regarded as a sum of single- and the two-photon transition amplitudes proportional to ε_1 and ε_2, where $\varepsilon_1 \sim (\mu_{1i}, g_k)$ and $\varepsilon_2 \sim q_b(k_1,k_2)$ or $q_s(k_1,k_2)$. Considering the situation when the two-photon amplitude is smaller than the single-photon amplitude $\varepsilon_2 < \varepsilon_1$, we conclude that beginning with the third-order term, the correlation diagrams (12) and (15), proportional to $\varepsilon_1^2\varepsilon_2$, can play an important role in the two-quantum decay process even for the two-atomic system consisted from one atom of each subsystems: D and Ξ (or D and V). For example, in the situation when $\varepsilon_1 = 0.7$ and $\varepsilon_2 = 0.25$, the magnitude of two-photon emission, $\varepsilon_2^2 = 0.0625$, becomes smaller than the cooperative magnitude $\varepsilon_1^2\varepsilon_2 = 0.1225$). In other words we can find the condition for which we can neglect

the decay rate of two-photon emission of the D atom in comparison with the cooperative effect described by expressions (12) and (15). This possibility to control the two-photon decay process of D atom with the decay process of Ξ or V excited three-level atom is given in the next section.

3. Two-photon energy transfer between the two three-level radiators

Master Eq. (10) can be used for the description of cooperative interaction between the dipole-forbidden and dipole-active radiators in two-photon exchanges. Indeed passing again from Schrodinger to Heisenberg pictures $Tr\left[\hat{W}(t)\hat{O}(0)\right] = Tr\left[\hat{W}(0)\hat{O}(t)\right]$, we can obtain from this expression the equation of the arbitrary atomic operator $\hat{O}(t)$. Let us firstly discuss the nonlinear interaction in which Ξ and D atoms enter in two-photon resonance as represented in **Figure 1A**. Studying the cooperative interaction between the dipole-forbidden and dipole-active radiators, the closed system of equations for the correlation functions can be found in such approach. Considering that the numbers of atoms in the each subsystem are relatively small, we can obtain the following generalized equation for the arbitrary operator O_b:

$$\frac{d\langle O_b(t)\rangle}{dt} = \frac{1}{2\tau_{\iota,1}} \sum_{l,j=1}^{N_\xi} \chi_1(j,l)\left\langle \hat{\Xi}^\iota_{1,l}(t)\left[\hat{O}_b(t), \hat{\Xi}^1_{\iota,j}(t)\right]\right\rangle$$

$$+ \frac{1}{2\tau_{\iota,2}} \sum_{l,j=1}^{N_\xi} \chi_2(j,l)\left\langle \hat{\Xi}^2_{\iota,l}(t)\left[\hat{O}_b(t), \hat{\Xi}^\iota_{2,j}(t)\right]\right\rangle$$

$$+ \frac{1}{2\tau_d} \sum_{l,j=1}^{N} \chi_d(j,l)\left\langle \hat{D}^+_l(t)\left[\hat{O}_b(t), \hat{D}^-_j(t)\right]\right\rangle$$

$$- \frac{i}{4\tau^b_{12d}} \sum_{m=1}^{N}\sum_{l=1}^{N_\xi}\sum_{j=1}^{N_\xi}\left\{ U_b(m,l,j)[\left\langle \hat{\Xi}^2_{\iota,l}(t)\hat{\Xi}^\iota_{1,j}(t)\left[\hat{O}_b(t), \hat{D}^-_m(t)\right]\right\rangle\right.$$

$$+ \left\langle \hat{\Xi}^\iota_{1j}(t)\hat{\Xi}^2_{\iota l}(t)\left[\hat{O}_b(t), \hat{D}^-_m(t)\right]\right\rangle] + U^*_b(j,l,m)\left\langle\left\{\left[\left[\hat{O}_b(t), \hat{\Xi}^\iota_{1,j}(t)\right], \hat{\Xi}^2_{\iota,l}(t)\right]\right.\right.$$

$$+ \left.\left.\left[\left[\hat{O}_b(t), \hat{\Xi}^2_{\iota,l}(t)\right], \hat{\Xi}^\iota_{1,j}(t)\right]\right\}\hat{D}^-_m\right\rangle\right\}$$

$$- \frac{i}{2\tau^b_{12d}} \sum_{m=1}^{N}\sum_{j=1}^{N_\xi}\sum_{l=1}^{N_\xi} V_b(j,m,l)\left\{\langle\hat{\Xi}^\iota_{1,j}(t)[\hat{\Xi}^2_{\iota,l}(t), \hat{O}_b(t)]\hat{D}^-_m(t)\rangle\right.$$

$$+ \langle\hat{\Xi}^2_{\iota,l}(t)[\hat{\Xi}^\iota_{1,j}(t), \hat{O}_b(t)]\hat{D}^-_m(t)\rangle\left.\right\} + H.c.$$

$$\tag{18}$$

In order to simplify this problem, we analyze below the situation in which we have only a single atom in each subsystem. In this case we can replace the operator O_b with the excitation numbers operators $\hat{N}_\alpha = \hat{\Xi}^\alpha_\alpha(t)$ and $\hat{N}_d = \hat{D}_z + 0.5$ of Ξ and D atoms, respectively. Here $\alpha = 1, 2$ and ι. When emission frequencies of the one-photon radiators coincide with $\omega_1 \simeq \omega_2 \simeq \omega_0$, the dependence (14) becomes real and positive defined function Ξ and D radiators. Here $[\exp[i\omega_0 r/c] - 1][\exp[-i\omega_0 r/c]$

$-1 = 2[1 - \cos{(\omega_0 r/c)}]$. According to this expression, the exchange integrals become

$$V_{12d}^b = \frac{2(1 - \cos{(2\pi r/\lambda_0)}}{(2\pi r/\lambda_0)^2}, \quad U_{12d}^b = \exp{[-i\omega_0 r/c]}V_{srd}.$$

In this case one can introduce the expression exchange rate $1/\tau_{srd}^b$ as a function of the distance between the dipole-active and dipole-forbidden subsystems:

$$\frac{1}{\tau_{12d}^b(r)} = \frac{1}{\tau_{srd}^b}\frac{2(1 - \cos{(2\pi r/\lambda_s)}}{(2\pi r/\lambda_s)^2}, \tag{19}$$

where $\lambda_0 = c/[2\pi\omega_0]$. Taking into account the above definitions and introducing the correlation functions between the polarizations of Ξ and D atoms $\langle \hat{F}_b(t,r) \rangle = i\left[\langle \hat{\Xi}_1^2(t)\hat{D}^-(t) \rangle - \langle \hat{D}^+(t)\hat{\Xi}_2^1(t) \rangle\right]$ and $\langle \hat{E}_b(t,x) \rangle = \langle \hat{\Xi}_1^2(t)\hat{D}^-(t) \rangle + \langle \hat{D}^+(t)\hat{\Xi}_2^1(t) \rangle$, we obtain the closed system of equations from expression (18):

$$\frac{d}{dt}\langle \hat{N}_2(t,x) \rangle = -\frac{\langle \hat{N}_2(t) \rangle}{\tau_{l,2}} - \frac{1}{4\tau_{12d}^b(x)}\left[\cos{(x)}\langle \hat{F}_b(t,x) \rangle - \sin{(x)}\langle \hat{E}(t,x) \rangle\right],$$

$$\frac{d}{dt}\langle \hat{N}_l(t,x) \rangle = \frac{\langle \hat{N}_2(t,x) \rangle}{\tau_{l,2}} - \frac{\langle \hat{N}_l(t,x) \rangle}{\tau_{l,1}} + \frac{1}{2\tau_{12d}^b(x)}\left[\cos{(x)}\langle \hat{F}_b(t,x) \rangle\right.$$
$$\left. - \sin{(x)}\langle \hat{E}(t,x) \rangle\right] + \frac{\langle \hat{F}_b(t,x) \rangle}{2\tau_{12d}^b(x)},$$

$$\frac{d}{dt}\langle \hat{N}_1(t,x) \rangle = \frac{\langle \hat{N}_l(t) \rangle}{\tau_{l,1}} - \frac{1}{4\tau_{12d}^b(x)}\left[\cos{(x)}\langle \hat{F}_b(t,x) \rangle - \sin{(x)}\langle \hat{E}_b(t,x) \rangle\right]$$
$$- \frac{1}{2\tau_{12d}^b(x)}\langle \hat{F}_b(t,x) \rangle;$$

$$\frac{d}{dt}\langle \hat{N}_d(t,x) \rangle = -\frac{\langle \hat{N}_d(t,x) \rangle}{\tau_d}$$
$$+ \frac{1}{4\tau_{12d}^b(x)}\left[\cos{(x)}\langle \hat{F}_b(t,x) \rangle + \sin{(x)}\langle \hat{E}_b(t,x) \rangle\right],$$

$$\frac{d}{dt}\langle \hat{F}_b(t,x) \rangle = -\frac{\langle \hat{F}_b(t,x) \rangle}{2}\left(\frac{1}{\tau_d} + \frac{1}{\tau_{l,2}}\right) \tag{20}$$
$$- \frac{1}{2\tau_{12d}^b(x)}\{\cos{(x)}[2\langle \hat{N}_2(t,x)\hat{N}_d(t) \rangle - \langle N_2(t,x) \rangle$$
$$- \langle \hat{N}_d(t)(\hat{N}_i(t) - \hat{N}_2(t)) \rangle + \langle \hat{N}_d(t)(1 - \hat{N}_2(t) - 2\hat{N}_i(t)) \rangle]$$
$$- 2\langle \hat{N}_d(t)\hat{N}_i(t) \rangle + 2\langle \hat{N}_d(t)\hat{N}_2(t) \rangle\};$$

$$\frac{d\langle \hat{E}_b(t,x) \rangle}{dt} = \frac{\langle \hat{E}_b(t,x) \rangle}{2}\left(\frac{1}{\tau_d} + \frac{1}{\tau_{l,2}}\right) - \frac{1}{2\tau_{12d}^b(x)}\sin{(x)}[2\langle \hat{N}_2(t)\hat{N}_d(t) \rangle$$
$$- \langle \hat{N}_2(t) \rangle + \langle \hat{N}_d(t)(\hat{N}_i(t) - \hat{N}_2(t)) \rangle$$
$$- \langle \hat{N}_d(t)(1 - \hat{N}_2(t) - 2\hat{N}_i(t)) \rangle];$$

$$\frac{d}{dt}\langle \hat{N}_2(t)\hat{N}_d(t) \rangle = -\langle \hat{N}_2(t)\hat{N}_d(t) \rangle\left[\frac{1}{\tau_{l,2}} + \frac{1}{\tau_d}\right],$$

$$\frac{d}{dt}\langle \hat{N}_l(t)\hat{N}_d(t) \rangle = \frac{\langle \hat{N}_2(t)\hat{N}_d(t) \rangle}{\tau_{l,2}} - \langle \hat{N}_l(t)\hat{N}_d(t) \rangle\left[\frac{1}{\tau_{l,1}} + \frac{1}{\tau_d}\right].$$

Using this system of Eq. (20), we can numerically study the cooperative nonlinear exchanges through the vacuum field between the Ξ and D radiators situated at relative distance x. One can observe that the spontaneous generation of photon pair by the D atom is drastically modified by the time increase of the cooperative correlation between the radiators. Indeed considering that the decay rate of the D atom $1/\tau_d$ is smaller than similar rates of the cascade transition in the Ξ atom ($\tau_d/\tau_{\xi,i} \simeq 6$; $\tau_d/(4\tau_{12d}) = 2$), we can numerically represent this dependence as a function of the relative time, t/τ_d, and the relative distance between the radiators, $x = 2\pi r/\lambda_0$. As shown in **Figure 4A**, the decay rate of D atom is drastically modified at small distances between the radiators which is in accordance with the analytic expressions (19). Considering that both atoms Ξ and D are prepared in the excited state, we observe the significant enhancement of the two-photon emission rate of the D radiator under the influence of the Ξ decay process.

Let us simplify the system of Eq. (20) in order to solve it exactly. Indeed, when dipole-active Ξ atom is situated at small distance relative to the D radiator ($x \ll 1$), the system of Eq. (20) is drastically simplified:

$$\frac{d}{dt}\langle \hat{N}_2(t)\rangle = -\frac{\langle \hat{N}_2(t)\rangle}{\tau_{i,2}} - \frac{\langle \hat{F}_b(t)\rangle}{4\tau_{12d}^b},$$

$$\frac{d}{dt}\langle \hat{N}_d(t)\rangle = -\frac{\langle \hat{N}_d(t)\rangle}{\tau_d} + \frac{1}{4\tau_{12d}^b}\langle \hat{F}_b(t)\rangle,$$

$$\frac{d}{dt}\langle \hat{F}_b(t)\rangle = -\frac{\langle \hat{F}_b(t)\rangle}{2}\left(\frac{1}{\tau_d} + \frac{1}{\tau_{i,2}}\right)$$

$$-\frac{1}{2\tau_{12d}^b}[4\langle \hat{N}_2(t)\hat{N}_d(t)\rangle + \langle \hat{N}_d(t)\rangle \qquad (21)$$

$$-\langle \hat{N}_2(t)\rangle - 5\langle \hat{N}_d\hat{N}_i\rangle],$$

$$\frac{d}{dt}\langle \hat{N}_2(t)\hat{N}_d(t)\rangle = -\langle \hat{N}_2(t)\hat{N}_d(t)\rangle\left[\frac{1}{\tau_{i,2}} + \frac{1}{\tau_d}\right],$$

$$\frac{d}{dt}\langle \hat{N}_i(t)\hat{N}_d(t)\rangle = \frac{\langle \hat{N}_2(t)\hat{N}_d(t)\rangle}{\tau_{i,2}} - \langle \hat{N}_i(t)\hat{N}_d(t)\rangle\left[\frac{1}{\tau_{i,1}} + \frac{1}{\tau_d}\right].$$

The exact solution of this linear system of equation can be represented through solution of characteristic equation $Y_\alpha = \sum_{j=1}^{5} C_\alpha^j \exp[\Theta_j t]$, where $\alpha = 1,2,3,4,5$ and $\{Y_\alpha\}$ are the atomic functions, $Y_1(t) = \langle \hat{N}_d(t)\rangle$, $Y_2(t) = \langle \hat{N}_2(t)\rangle$, $Y_3(t) = \langle \hat{F}_b(t)\rangle$, $Y_4(t) = \langle\langle \hat{N}_2(t)\hat{N}_d(t)\rangle\rangle$, and $Y_5(t) = \langle\langle \hat{N}_i(t)\hat{N}_d(t)\rangle\rangle$; the solution of characteristic equation is

$$\Theta_1 = -\left(\frac{1}{\tau_2} + \frac{1}{\tau_d}\right); \quad \Theta_2 = -\left(\frac{1}{\tau_1} + \frac{1}{\tau_d}\right); \quad \Theta_3 = -\frac{1}{2}\left(\frac{1}{\tau_d} + \frac{1}{\tau_{i,2}}\right);$$

$$\Theta_{4,5} = -\frac{1}{2}\left\{\frac{1}{\tau_{i,2}} + \frac{1}{\tau_d} \pm \sqrt{\left(\frac{1}{\tau_d} - \frac{1}{\tau_{i,2}}\right)^2 - \frac{1}{\tau_{12b}^2}}\right\}. \qquad (22)$$

The coefficients $\{C_\alpha^j\}$ are determined from the initial conditions. As follows from the numerical estimation plotted in **Figure 4B** and solutions of characteristic
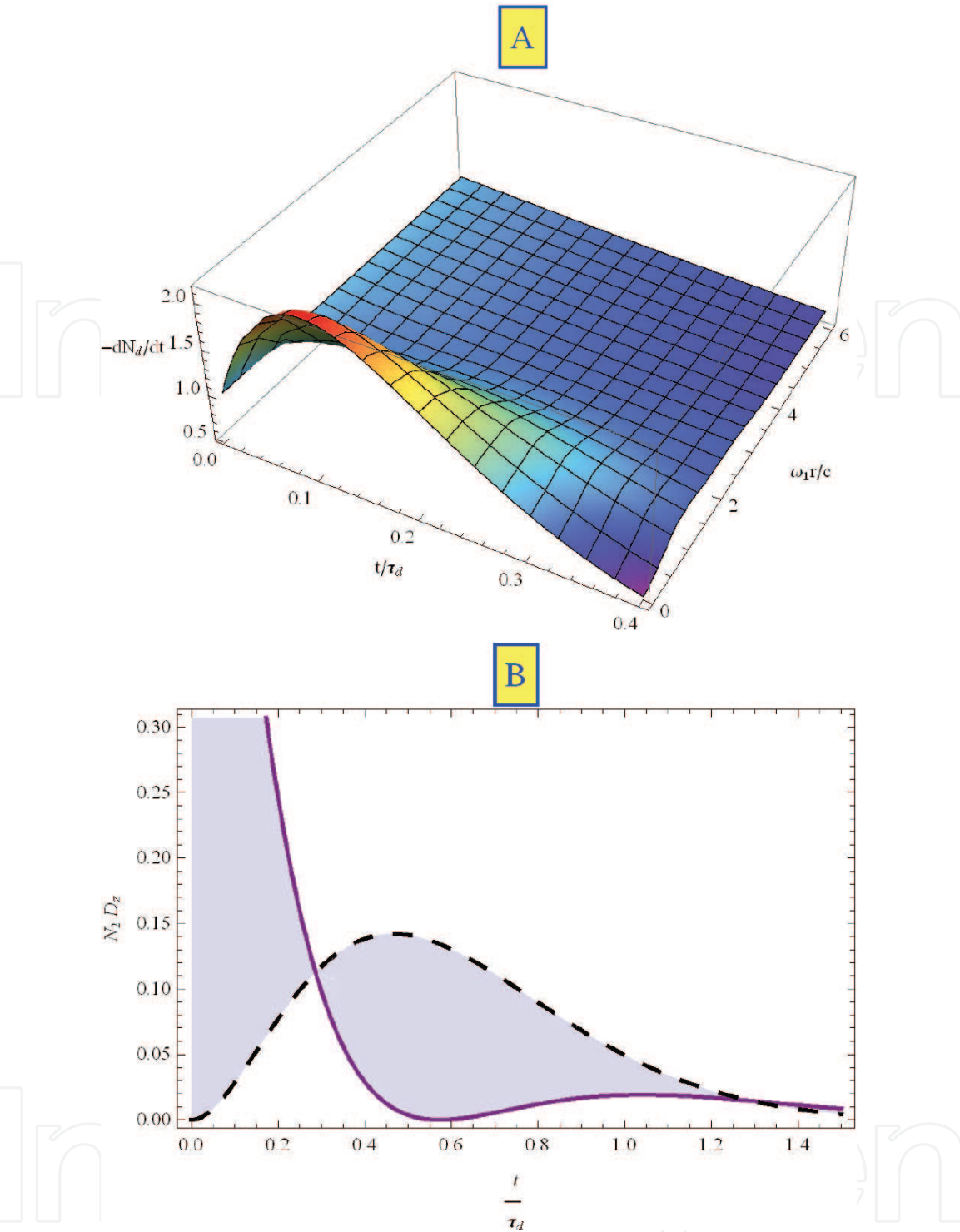
Figure 4.
(A) The decay rate $-d\langle N_d/dt \rangle$ of the dipole-forbidden transitions of the D radiator under the influence of Ξ three-level radiator. This solution of Eq. (21) is plotted as function of t/τ_d and relative distance between the radiators $x = \omega_1 r/c$, for the following parameters of the system: $\langle N_1 \rangle = \langle N_i \rangle = 0., \langle N_2 \rangle = 1., \langle N_d \rangle = 1,$ $\tau_d/\tau_1 = \tau_d/\tau_2 = 6,$ and $\tau_d/(4\tau_{12d}) = 2.$ (B) The decay process of excited state $|2\rangle$ of three-level system (thick line) and the transfer of the excitation from the Ξ radiator to D atom (dashed line) in the process of cascade emission of Ξ atom situated at relative distance $x < < 1$ for the same parameters of the system and excitation conditions: $\langle N_1 \rangle = \langle N_i \rangle = 0, \langle N_2 \rangle = 1.,$ and $\langle N_d \rangle = 0.$

in Eq. (22), the oscillatory decay of the atomic inversion is possible, when $1/\tau_d = 1/\tau_{i,2}$. In this case the solutions $\Theta_{4,5}$ become complex. We observe such an oscillation of the atoms inversion of Ξ radiator prepared initially in the excited state. In this process the rate of energy transfer from Ξ to D atoms represented in **Figure 4B** has the oscillator behavior. In the case of the excitation of D, the coupling between the radiators becomes more effective, when the virtual level of

the D atom is situated between the excited and ground states (see **Figure 4B**). As the virtual states of the D radiator is off from the resonance with the dipole-active transitions of the Ξ radiators, the excitation of D atom takes place only with the absorption of both emitted photons by the Ξ atom. The cooperative effects between the Ξ and D radiators are described by second-order correlation function $G_2 = \left\langle \hat{E}^-(t)\hat{E}^-(t)\hat{E}^+(t)\hat{E}^+(t) \right\rangle = G_2^0 + \alpha\langle \hat{F}_b(t) \rangle$. Here G_2^0 was derived in Ref. [5]. The contribution to the second-order correlation function remains larger than the square value of the first-order correlation function $G_1 = \langle E^-(t)E^+(t) \rangle$, so that we can conclude that new cooperative effects between single- and two-photon transitions of D and Ξ subsystems play an important role in the two-photon decay process. Let us now return to the V three-level system in scattering interaction with the D system as this is represented in **Figure 1B**. In accordance with master Eq. (10) and its analytic representation (15), we can obtain the following expression for arbitrary atomic operators $\hat{O}_s(t)$.

$$
\begin{aligned}
\frac{d\left\langle \hat{O}_s(t) \right\rangle}{dt} &= \frac{1}{2\tau_{\iota,1}} \sum_{l,j=1}^{N_v} \chi_a(j,l) \left\langle \hat{V}_{\iota,l}^1(t) \left[\hat{O}^{(s)}(t), \hat{V}_{1,j}^\iota(t) \right] \right\rangle \\
&+ \frac{1}{2\tau_{\iota,2}} \sum_{l,j=1}^{N_v} \chi_s(j,l) \left\langle \hat{V}_{\iota,l}^2(t) \left[\hat{O}^{(s)}(t), \hat{V}_{2,j}^\iota(t) \right] \right\rangle \\
&+ \frac{1}{2\tau_d} \sum_{l,j=1}^{N} \chi_d(j,l) \left\langle \hat{D}_l^+(t) \left[\hat{O}^{(s)}(t), \hat{D}_j^-(t) \right] \right\rangle \\
&- \frac{i}{2\tau_{sad}^s} \sum_{m=1}^{N} \sum_{j=1}^{N_v} \sum_{l=1}^{N_v} V_s(j,m,l) \left\langle \hat{V}_{\iota,j}^1(t) \left[\hat{O}^{(s)}(t), \hat{D}_m^-(t) \right] \hat{V}_{2,l}^\iota(t) \right\rangle \\
&+ \frac{i}{2\tau_{sad}^s} \sum_{m,j,l=1} \left\{ U_s(j,m,l) \langle [\hat{O}^{(s)}(t), \hat{V}_{\iota,j}^1(t)] \hat{V}_{2l}^\iota(t) \hat{D}_m^-(t) \rangle \right. \\
&\left. + U_s^*(j,m,l) \langle \hat{V}_{\iota,j}^1(t) \hat{D}_m^-(t) [\hat{O}^{(s)}(t), \hat{V}_{2,l}^\iota(t)] \rangle \right\} + H.c.
\end{aligned}
\tag{23}
$$

The similar expression can be obtained for a Λ three-level system in interaction with D radiators, doing the substitution $\hat{V}_{\alpha,j}^\beta \rightarrow \hat{\Lambda}_{\beta j}^\alpha$. For two atoms in each subsystem, an attractive peculiarity follows from this substitution. If $O_s(t)$ is the inversion of the D atom, the direct modification of the D atomic excitation by Λ three-level atom is equal to zero $\left\langle \hat{\Lambda}_{1,l}^\iota(t) \hat{\Lambda}_{\iota,l}^2(t) \left[\hat{N}_d(t), \hat{D}_m^-(t) \right] \right\rangle = 0$ due to the operator product $\hat{\Lambda}_{1,l}^\iota(t) \hat{\Lambda}_{\iota,l}^2(t) = 0$ for the same atom. In order to obtain the closed system of equation from master Eqs. (15) and (23), we consider the simple interaction of two atoms in the scattering process represented by the analytical scheme of **Figure 1B**. In this case we introduce the new indexes "s" and "a" instead of "1" and "2", which correspond to the Stokes and anti-Stokes scattering frequencies ω_s and ω_a. Considering that the anti-Stokes frequency ω_a is larger than Stokes ω_s, one can approximate the exchange integrals (17) with expression

$$
V_s \simeq \frac{\sin(x_a)}{x_a} + i \frac{1 - \cos(x_a)}{x_a}.
\tag{24}
$$

Here $x_a = \omega_a r/c$. The mean values of the operators $\langle \hat{N}_s \rangle = \left\langle \hat{V}_2^2 \right\rangle$, $\langle \hat{N}_a \rangle = \left\langle \hat{V}_1^1 \right\rangle$, and $\langle \hat{N}_d \rangle$ are considered the populations of excited states of V and D radiators,
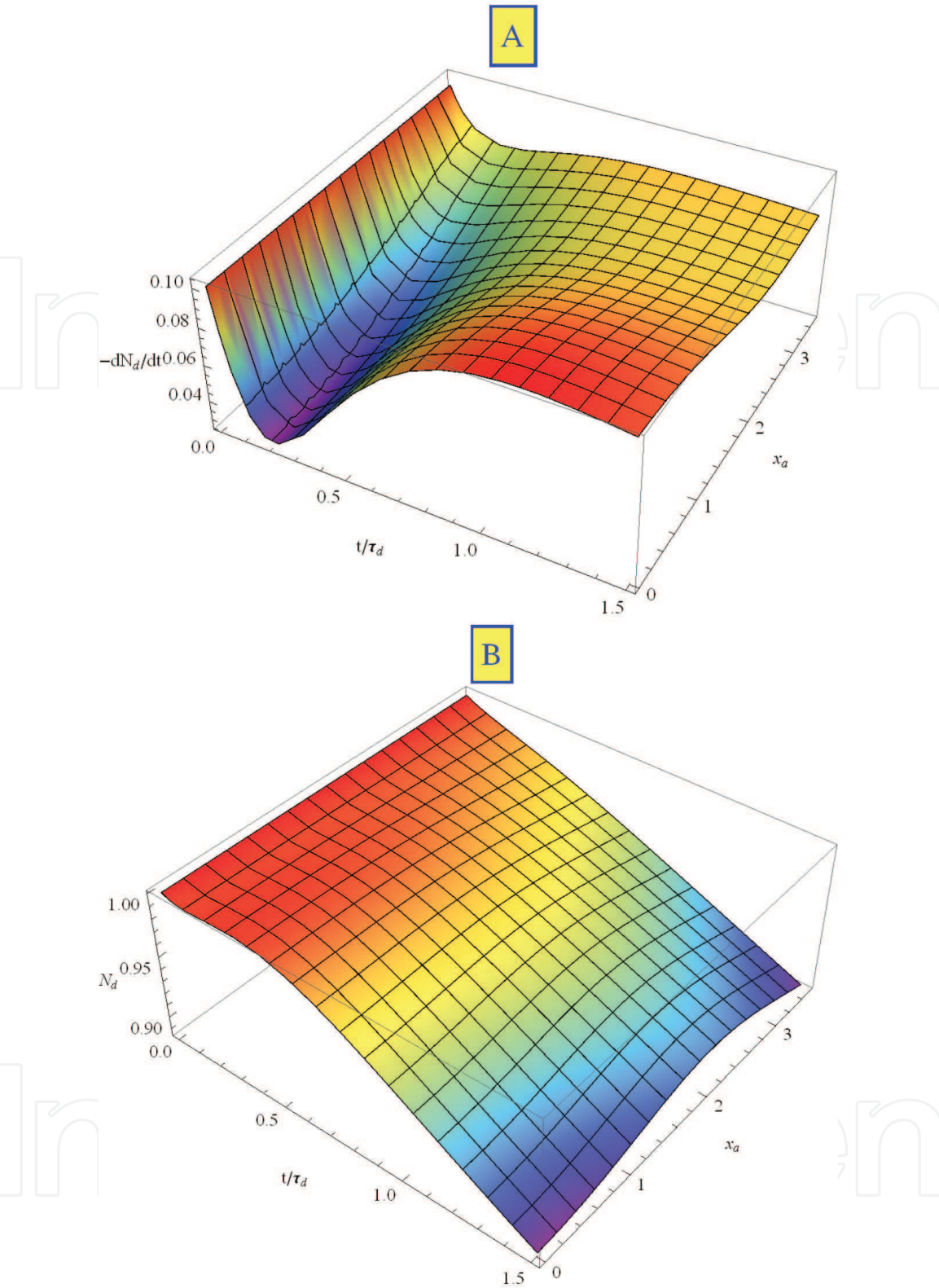
Figure 5.
The decay process of the dipole-forbidden transitions of the D radiator under the influence of V three-level radiator for following parameter atom for following parameters of the system, $\langle N_a \rangle = 0.5$, $\langle N_s \rangle = 0.5$, $\langle N_d \rangle = 1$, $\tau_a/\tau_d = 0.1$, $\tau_a/\tau_s = 6$, and τ_a/τ_{asd}, (A) represents the decay rate $-d\langle N_d/dt \rangle$ and (B) represents the excitation of the D atom plotted as the numerical solution of the system of Eq. (25) as function of t/τ_a) and relative distance $x_a = 2\pi r/\lambda_a$ in the three-dimensional representation.

respectively. The functions $\langle \hat{F}_s(t, x_a) \rangle = i \left[\left\langle \hat{V}_2^1(t) \hat{D}^-(t) \right\rangle - \left\langle \hat{D}^+(t) \hat{V}_1^2(t) \right\rangle \right]$,

$\langle \hat{E}_s(t, x_a) \rangle = \left[\left\langle \hat{V}_2^1(t) \hat{D}^-(t) \right\rangle - \left\langle \hat{D}^+(t) \hat{V}_1^2(t) \right\rangle \right]$, $\langle \hat{N}_d \hat{N}_s \rangle$, and $\langle \hat{N}_d \hat{N}_a \rangle$ describe the

polarization and population correlations between the atoms Ξ and D. For this two-atom system, we can obtain the following closed system of equations from generalized equation (23).

$$\frac{d}{dt}\langle\hat{N}_\iota(t,x_a)\rangle = \frac{1}{\tau_{\iota,a}}\langle\hat{N}_a(t,x_a)\rangle + \frac{1}{\tau_{\iota,s}}\langle\hat{N}_s(t,x_a)\rangle + \frac{1}{\tau_{sad}^s}\frac{1-\cos(x_a)}{x_a}\langle\hat{E}_s(t,x_a)\rangle,$$

$$\frac{d}{dt}\langle\hat{N}_a(t,x_a)\rangle = -\frac{1}{\tau_{\iota,a}}\langle\hat{N}_a(t,x_a)\rangle$$

$$+ \frac{1}{2\tau_{sad}^s}\left[\frac{\sin(x_a)}{x_a}\langle\hat{F}_s(t,x_a)\rangle - \frac{1-\cos(x_a)}{x_a}\langle\hat{E}_s(t,x_a)\rangle\right],$$

$$\frac{d}{dt}\langle\hat{N}_s(t,x_a)\rangle = -\frac{1}{\tau_{\iota,s}}\langle\hat{N}_s(t,x_a)\rangle$$

$$- \frac{1}{2\tau_{sad}^s}\left[\frac{\sin(x_a)}{x_a}\langle\hat{F}_s(t,x_a)\rangle + \frac{1-\cos(x_a)}{x_a}\langle\hat{E}_s(t,x_a)\rangle\right],$$

$$\frac{d}{dt}\langle\hat{N}_d(t,x_a)\rangle = -\frac{1}{\tau_d}\langle\hat{N}_d(t,x_a)\rangle$$

$$+ \frac{1}{2\tau_{sad}^s}\left[\frac{\sin(x_a)}{x_a}\langle\hat{F}_s(t,x_a)\rangle - \frac{1-\cos(x_a)}{x_a}\langle\hat{E}_s(t,x_a)\rangle\right],$$

$$\frac{d}{dt}\langle\hat{F}_s(t,x_a)\rangle = -\frac{1}{2}\left\{\frac{1}{\tau_d}+\frac{1}{\tau_{\iota,s}}+\frac{1}{\tau_{\iota,a}}\right\}\langle\hat{F}_s(t,x_a)\rangle$$

$$+ \frac{1}{\tau_{sad}^s}\frac{\sin(x_a)}{x_a}\left[\langle\hat{N}_d(t,x_a)\hat{N}_s(t,x_a)\rangle - \langle(1-\hat{N}_d(t,x_a))\hat{N}_a(t,x_a)\rangle\right],$$

$$\frac{d}{dt}\langle\hat{E}_s(t,x_a)\rangle = -\frac{1}{2}\left\{\frac{1}{\tau_d}+\frac{1}{\tau_{\iota,s}}+\frac{1}{\tau_{\iota,a}}\right\}\langle\hat{E}_s(t,x_a)\rangle$$

$$- \frac{1-\cos(x_a)}{x_a\tau_{sad}^s}\left\{\langle\hat{N}_d(t,x_a)\hat{N}_s(t,x_a)\rangle + \langle(1-\hat{N}_d(t,x_a))\hat{N}_a(t,x_a)\rangle\right\}$$

$$\frac{d}{dt}\langle\hat{N}_d(t,x_a)\hat{N}_s(t,x_a)\rangle = -\left[\frac{1}{\tau_d}+\frac{1}{\tau_{\iota,s}}\right]\langle\hat{N}_d(t,x_a)\hat{N}_s(t,x_a)\rangle$$

$$- \frac{1}{2\tau_{sad}^s}\left[\frac{\sin(x_a)}{x_a}\langle\hat{F}_s(t,x_a)\rangle + \frac{1-\cos(x_a)}{x_a}\langle\hat{E}_s(t,x_a)\rangle\right],$$

$$\frac{d}{dt}\langle\hat{N}_d(t,x_a)\hat{N}_a(t,x_a)\rangle = -\left[\frac{1}{\tau_d}+\frac{1}{\tau_{\iota a}}\right]\langle\hat{N}_d(t,x_a)\hat{N}_a(t,x_a)\rangle.$$

$$(25)$$

As follows from the system (25), and numerical simulation plotted in **Figure 5** the first $\langle\hat{N}_d\rangle/\tau_d$ and second terms $(1/\tau_{sad}^s)\langle\hat{F}_s\rangle$ describe the generation rate of entangled photon pairs and scattering rate with absorption of Stokes photon and generation of two anti-Stokes photons by the system formed from V and D atoms. When the time tends to infinity, all excited atomic energies $E_0 = \hbar\omega_a + \hbar\omega_s + \hbar\omega_d$ of three-level V and two-level D atoms are emitted by the system. Taking into account the conservation law in the scattering process $\omega_a - \omega_s - \omega_d = 0$, we observe that this cooperation between the atoms becomes predominant, when the collective scattering rate $1/\tau_{sad}^s$ increases. In other words, the probability of absorption of Stokes photon $\hbar\omega_s$ which is accompanied with the generation of the new anti-Stokes photon $\hbar\omega_a$ by D atom becomes possible. In this case two atoms represented in the **Figure 1B** can generate an entangled anti-Stokes photons with energy $E_0 = 2\hbar\omega_a$. The possibility of the excitation transfer between the atoms Ξ and D represented in **Figure 4B** can be found in the special preparation of the system.

We can conclude that it is possible to study all cooperations two-photon process between single atoms in each system represented in **Figure 1A–C**. For example, the system of Eqs. (20) and (25) can be solved simultaneously taking into consideration scattering and two-photon transitions. In this case the effective energy transfer of the excitation between the atoms Ξ, V, and D radiator prepared in the special initial states can open the new possibilities of non-resonance interaction between the atomic subsystems.

4. Conclusions

This chapter proposed the cooperative effects between three-level system and dipole-forbidden two-level systems in nonlinear interaction through the vacuum field during the spontaneous emission time. The possibility of cooperative migration of energy from one excited dipole-active three-level atom to another takes place with phase retardation effects and depends on the position of atoms in the system. This excitation transfer from dipole-active to dipole-forbidden subsystems takes place with phase dependence amplitudes, so that the cooperative excitation of the system consisted from two species of atoms depends on the retardation of radiation along the sample and geometry of the system. This follows from the excited or ground state of one of the radiators represented in **Figures 4** and **5**. As in Ref. [20], the exchanges between the Ξ (or V) three-level atom and D take place with the absorption and emission of two quanta, but in this chapter, we take into consideration the real and imaginary parts of exchange integrals. In this case, two correlation functions introduced functions $\langle \hat{F}_{b(s)}(t, x_a) \rangle$ and $\langle \hat{E}_{b(s)}(t, x_a) \rangle$, which modify the dynamics of possible excitation of D atoms by Ξ and V radiators. The scattering transfer of the energy between the excited state of V three-level radiator and dipole-forbidden transitions of D two-level atoms are effective when the dipole-forbidden atom enters in the two-photon resonance with the energy difference between the two dipole transitions (**Figures 1A** and **5A**). When the atom D is in the excited state, the emitted Stokes photon by one atom of the V systems can be absorbed by another radiator from the D subsystem, so that two radiators pass into the ground state generating two anti-Stokes photons with energies $E_0 = 2\hbar\omega_a$. The opposite situation can be observed when D atom is prepared in the ground state.

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