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Modeling of the dynamics of selective resonant fluorescence of two interacting nanoparticles

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A system of self-consistent differential equations of the first order in time is presented, which reflects the dynamics of resonant fluorescence of two interacting two-level particles when one of them is irradiated with a long rectangular pulse of light. This system of equations is obtained based on the analysis of equations describing the dynamics of the states of a common composite system of a marked pair of particles and a quantized radiation field, taking into account the continuous absorption of a fluorescence photon by the macroscopic environment of particles, which is attributed to the radiation of one or another of the interacting particles when its ground state is populated („selective" fluorescence). Along with the dynamics of the population of the initial collective state of the general system, the dynamics of the population and coherence of the main and excited collective states of particles when they absorb one of the irradiation photons, the resulting system describes the dynamics of the population of each particle in its ground state, recorded with the absorption of a resonant fluorescence photon noted above. This description of the dynamics of the population of the ground state of each of the particles distinguishes the above system of equations from systems that, in relation to the fluorescence of this pair of particles, describe the dynamics of their collective ground state, such as, for example, the system of optical Bloch equations. Accordingly, the characteristic features of the dynamics of selective resonant fluorescence under consideration differ significantly from the characteristic features of the dynamics of resonant fluorescence described by the system of optical Bloch equations. These differences are illustrated by figures reflecting the results of numerical solutions of the reduced system of equations and solutions of the corresponding system of optical Bloch equations.

Keywords: mathematical modeling, interacting two-level nanoparticles, dynamics of resonant fluorescence.

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Introduction

In the theoretical and experimental study of various aspects of resonant fluorescence (RF) of two interacting atoms, ions, and semiconductor quantum dots, modeling of the dynamics of states of interacting two-level nanoparticles irradiated by light is successfully used [1,2]. Typically, this modeling is based on the application of classical radiation field theory and consideration of a pair of particles as a closed system when describing the dynamics of their collective states by a governing equation for the density matrix, for example, a system of optical Bloch equations (OBE). As noted in [3],„the density matrix description of physical objects can be considered as the special case of an even more general development based upon the analysis of experimental situations realized by selective measurements". The formalism corresponding to this method for modeling the population dynamics of the states of two different two-level particles coupled by dynamic interaction and the spectral characteristics of their spontaneous fluorescence (SF) is given in [4,5]. This formalism is based on the use of the theory of selective measurements of the population dynamics of the ground state of one of the particles when an SF photon is absorbed by this particle. It was taken into account that the environment of the particles (the apparatus [3]) separates the ensemble of the particle pair into subensembles with selection of a subensemble of one particle and „rejecting all others" [3]. Accordingly, in [4,5] the separation of the Hilbert space of states of a general composite system into subspaces of states related to the states of one of the particles and the SF photon emitted by this particle is used. At the same time, the system of Schrĕdinger equations for amplitudes of the states of these subspaces is written in such a form that solutions to it allowed obtaining expressions for the probability of emission of a SF photon by one or another particle when registering its ground state (selective SF of two particles). The corresponding expressions for the population of the ground state of each particle, the population and coherence of the excited collective states of particles and their time derivatives were obtained in analytical form in [4]. The equations connecting these expressions are written in the form of a system of equations for the dynamics of population and coherence of particle states under their selective SF.

This study applies such an approach to obtain a system of equations for the dynamics of the population and coherence of the states of two particles under their selective RF, excited by irradiation of one of the particles with a long rectangular

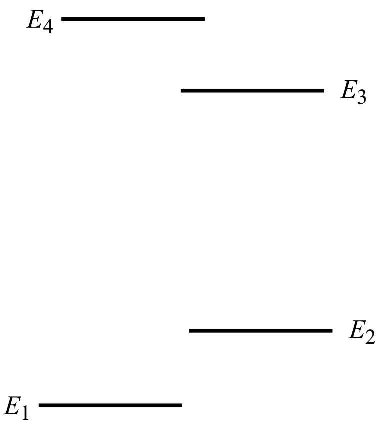

Figure 1. Diagram of the relative positions of particle energy levels.

pulse of light with a carrier frequency close to its eigen transition frequency.

Model and equations of a composite system

Let us write as follows the Hamiltonian of a system of closely spaced two „point" stationary two-level particles (particle A and particle B), which interact with each other and with the quantized radiation field [6]:

$$H = H_f + W + \sum_{j=A,B} H_j + V_j,$$

where H_j is Hamiltonian of the radiation field; W is energy operator of the dynamic interaction between particles, $H_A(H_B)$ is Hamiltonian of particle $A(B)$, $V_A = -(\mathrm{ed}_A)$ is energy operator of the electrodipole (d_A being operator of the dipole moment of particle A) interaction of particle A with the transverse components of the radiation field and a similar expression $V_B = -(\mathrm{ed}_B)$ for particle B. The orientation of the particle transition dipoles and the direction of irradiation are such that it excites only particle A. The distance between particles is much less than the wavelength of the SF of a free particle and the wavelength of the SF of free particle B.

Let us introduce the following notation: $|n\rangle$ ($n = 1,\ 4$) are eigenstates of particle A with energies E_1 and E_4; $|n\rangle$ ($n = 2,\ 3$) are eigenstates of particle B with energies E_2 and E_3 (for definiteness we assume $E_4 \geq E_3$ and $E_2 \geq E_1$); $\hbar\omega_{nm} = (E_n - E_m)$, where $n = 4, 3, 2$; $m = 3, 2, 1$; $\omega_{41} - \omega_{32} = \omega_{43} + \omega_{21} \equiv 2\Delta$, where $0 < \Delta \ll \omega_{41}, \omega_{32}$. Fig. 1 schematically shows the relative position of energy levels E_n shown with segments of horizontal lines with approximately equal differences $E_4 - E_3 > 0$ and $E_2 - E_1 > 0$ (as is often assumed in modeling the dynamics of states of interacting ions, atoms [2]).

Let us denote the initial state of the irradiation field as $|\lambda\rangle$. This field contains an integer number $N \geq 1$ of photons λ with frequency ω_L from the frequency range meeting the condition of $\omega_{41} \gg |\omega_L - \omega_{41}|$; here λ is a set of indices characterizing the wave vector of the photon and the state of its polarization. Let us denote the state of the field containing $N - 1$ photons λ as $|0\rangle$.

Let us consider the following orthonormalized states as the basis states of the composite system: initial state $|12\lambda\rangle$, two „intermediate" excited states $|420\rangle$ and $|130\rangle$, and states $|12\sigma\rangle$, $|12\nu\rangle$, which [4–6] are used to take into account the information on „what is the radiation path" [7] of photon fluorescence: one or another of the interacting particles when its ground state is populated (by the „Heisenberg jump", as noted on page 125 in [8]) as a result of the absorption of this photon by the „corresponding particle".

For the volume L^3 of space where the particles and the radiation field are located, let us denote the matrix element of the operator V_A for the states $|12\lambda\rangle$ and $|420\rangle$ as:

$$\langle 12\lambda|V_A|420\rangle = iL^{-3/2}\sqrt{2\pi N\hbar\omega_L}(d_\lambda)_{14} \equiv V^{40}_{1\lambda} = i\hbar\Omega_\lambda,$$

where $(d_\lambda)_{14}$ is matrix element for states $|1\rangle$ and $|4\rangle$ of the projection of the operator d on the direction of photon polarization λ, Ω_λ is Rabi frequency. Here, to simplify the notation, subscripts „2"are omitted in $V^{420}_{12\nu}$ and then in $V^{420}_{12\sigma}$, also subscripts „1"are omitted in $V^{130}_{12\nu}$ in the following text. Let us denote the matrix elements in the same way:

$$\langle 12\sigma|V_A|420\rangle = iL^{-3/2}\sqrt{2\pi\cdot\hbar\omega_\sigma}\cdot(d_\sigma)_{14} \equiv V^{40}_{1\sigma},$$

$$\langle 12\nu|V_B|130\rangle = iL^{-3/2}\sqrt{2\pi\cdot\hbar\omega_\nu}\cdot(d_\nu)_{23} \equiv V^{30}_{2\nu}.$$

The W will be meant as an operator of energy of the dipole-dipole interaction of particles, as, for example, in [9] when describing the interaction of two alkaline-earth atoms closely spaced from each other or when describing the interaction of a pair of CdSe/ZnS quantum dots [10], which relative position geometry was determined from their RF. For the matrix elements of the operator W, the following notation is used: $\langle 42|W|13\rangle = \langle 13|W42\rangle| \equiv \hbar w$.

Schrëdinger equations for the amplitudes $b_k(t)$ of the eigenstates $|k\rangle$ of the introduced Hamiltonian in the interaction representation have the following form:

$$i\hbar\dot{b}_k(t) = \sum_l (V_A + V_B + W)_{kl} b_l(t)$$

$$\times \exp[i(E_k - E_l)t/\hbar] + i\hbar\delta_{kl}\delta(t),$$

where i, k, l is set of subscripts denoting the basic orthonormalized states of the general system; δ_{ki} is Kronecker delta ($\delta_{kl} = 0$ for $k \neq i$ and $\delta_{ki} = 1$ for $k = i$). The introduction of an inhomogeneous term $i\hbar\delta_{ki}\delta(t)$ to preserve the normalization of amplitudes $b_k(t)$ for time values of both $t > 0$ and $t < 0$, including $t = 0$, is discussed, for example, in [11]. The use of the above matrix elements of the operator of particles interaction and the radiation field corresponds to the description of the dynamics of states of a composite system, taking into account transitions with conservation of its energy, neglecting the small contribution to the amplitude of these states from virtual transitions which do not conserve the energy of the system (the resonance approximation).

Bearing in mind what was stated in the Introduction, let us write the system of Schrëdinger equations in such a way that it reflects the initial condition $b_{12\lambda}(t=0)=1$ and the absence of interference between the states $|12\lambda\rangle$, $|12\sigma\rangle$, and $|12\nu\rangle$. This system has the following form:

$$i\hbar\dot{b}_{12\lambda}(t)=V_{1\lambda}^{40}b_{420}(t)\cdot\exp(i\bar{\omega}_\lambda t)+i\hbar\delta(t),$$

$$i\hbar\dot{b}_{420}(t)=V_{40}^{1\lambda}b_{12\lambda}(t)\cdot\exp(-i\bar{\omega}_\lambda t)+\sum_\sigma V_{40}^{1\sigma}b_{12\sigma}(t)$$

$$\times\exp(-i\bar{\omega}_\sigma t)+\hbar w b_{130}(t)\cdot\exp(i2\Delta t),$$

$$i\hbar\dot{b}_{130}(t)=\sum_\nu V_{30}^{2\nu}b_{12\nu}(t)\cdot\exp[-i(\bar{\omega}_\nu+2\Delta)t]$$

$$+\hbar w b_{420}(t)\cdot\exp(-i2\Delta t),$$

$$i\hbar\dot{b}_{12\sigma}(t)=V_{1\sigma}^{40}b_{420}(t)\cdot\exp(i\bar{\omega}_\sigma t),$$

$$i\hbar\dot{b}_{12\nu}t=V_{2\nu}^{30}b_{130}(t)\cdot\exp[i(\bar{\omega}_\nu+2\Delta)t],$$

where $\bar{\omega}_\lambda=\omega_\lambda-\omega_{41}$, $\bar{\omega}_\sigma=\omega_\sigma-\omega_{41}$, $\bar{\omega}_\nu=\omega_\nu-\omega_{41}$.

As noted in the Introduction, in [4,5] solutions to the system of Schrëdinger equations were found in an analytical form, reflecting the absence of interference between the states $|12\sigma\rangle$ and $|12\nu\rangle$ when considering the SF of a pair of particles, when at the initial moment of time particle A was excited. By using these solutions in [4], expressions for the population and coherence of particle states were obtained in an analytical form and the relationships between their time derivatives were determined, the set of which looks like a system of strictly defined equations describing the dynamics of particle states under SF. For example, if the following notations are used:

$$\sum_\sigma|b_{12\sigma}(t)|^2\equiv\rho_{11}(t),\qquad |b_{420}(t)|^2\equiv\rho_{44}(t),$$

$$\sum_\nu|b_{12\nu}(t)|^2\equiv\rho_{22}(t),\qquad |b_{130}(t)|^2\equiv\rho_{33}(t),$$

(which will be adopted in the following text), then the equation obtained in [4] for the time derivative of the population of the ground state of particle A and the population of its excited state is as follows:

$$\dot{\rho}_{11}(t)=2\gamma_4\rho_{44}(t),$$

where

$$\gamma_4=\pi\hbar^{-1}\sum_\sigma|V_{1\sigma}^{40}|^2\delta(E_{40}-E_{1\sigma})=2\omega_{41}^3d_{41}^2/3\hbar c^3$$

is radiative decay constant of the excited state of particle A [11]. In the same way, for particle B the following is obtained:

$$\dot{\rho}_{22}(t)=2\gamma_3\rho_{33}(t),$$

where

$$\gamma_3=\pi\hbar^{-1}\sum_\nu|V_{2\nu}^{30}|^2\delta(E_{30}-E_{2\nu})=2\omega_{32}^3d_{32}^2/3\hbar c^3$$

is radiative decay constant of the excited state of particle B (it is assumed that $\gamma_4\ll\omega_{41}$ and $\gamma_3\ll\omega_{32}$).

When using Fourier representations

$$b_k(t)=i(2\pi)^{-1}\int_{-\infty}^{+\infty}G_{ki}(E)\cdot\exp[i(E_k-E)t/\hbar]dE,$$

$$i\hbar\delta(t)=i(2\pi)^{-1}\int_{-\infty}^{+\infty}\exp[i(E_k-E)t/\hbar]dE,$$

the $G_{ki}(E)$ matrix is determined by solving the system of equations

$$(E-E_k)\cdot G_{ki}(E)=\sum_l(V_A+V_B+W)_{ki}G_{li}(E)+\delta_{ki}.$$

With the subscript i and the dependence G_{ki} on E omitted for simplicity, the system of equations for the selective RF under consideration can be written in the following form:

$$(E-E_{12\lambda})\cdot G_{12\lambda}=V_{1\lambda}^{40}G_{420}+1,$$

$$(E-E_{420})\cdot G_{420}=V_{40}^{1\lambda}G_{12\lambda}+\sum_\sigma V_{40}^{1\sigma}G_{12\sigma}+\hbar wG_{130},$$

$$(E-E_{130})\cdot G_{130}=\sum_\nu V_{30}^{2\nu}G_{12\nu}+\hbar wG_{420},$$

$$(E-E_{12\sigma})\cdot G_{12\sigma}=V_{1\sigma}^{40}G_{420}.$$

$$(E-E_{12\nu})\cdot G_{12\nu}=V_{2\nu}^{30}G_{130}.$$

According to the replacement assumed in [11] for the dividing the left-hand side of the equations of this system by $E-E_k$ with the multiplying their right-hand side by $\xi(E-E_k)$, the following notations are used:

$$i\hbar\gamma_4=-\sum_\sigma|V_{1\sigma}^{40}|^2\xi(E-E_{1\sigma}),$$

$$i\hbar\gamma_3=-\sum_\nu|V_{2\nu}^{30}|^3\xi(E-E_{2\nu}),$$

where

$$\xi(E)=P/E-i\pi\delta(E),$$

P/E is main value of the function $1/E$. In the following text [11] the expressions γ_m ($m=4$, 3) will be meant as the above-mentioned constants γ_4 and γ_3. Using these notations, the following solutions are yielded:

$$G_{12\lambda}(E)=[E_IE_{II}-(\hbar w)^2]/F(E),$$

$$G_{420}(E)=V_{40}^{1\lambda}E_I/F(E),$$

$$G_{130}(E)=V_{40}^{1\lambda}\hbar w/F(E),$$

$$G_{12\sigma}(E)=V_{12\lambda}^{420}V_{12\sigma}^{420}E_{II}\xi(E-E_{12\sigma})/F(E),$$

$$G_{12\nu}(E) = V^{420}_{12\lambda}V^{130}_{12\nu}\hbar w\xi(E - E_{12\nu})/F(E),$$
$$F(E) = E_0(E_I E_{II} - \hbar^2 w^2) - \hbar^2\Omega^2 E_I,$$
$$E_0 = E - E_{12\lambda}, \qquad E = E - E_{130} + i\hbar\gamma_3,$$
$$E_{II} = E - E_{420} + i\hbar\gamma_4.$$

Based on the form of the resulted functions $G_k(E)$, a conclusion can be made that finding a solution $b_k(t)$ to the above system of equations in analytical form (as a function of particle parameters and the irradiation field) seems to be a rather difficult task. Thus, the calculating them as the inverse Fourier representation of functions $G_k(E)$ using the theory of residues requires knowledge of the roots of a cubic (see function $F(E)$) algebraic equation with complex coefficients (not a quadratic equation as in the calculations in [4,5]). Similar difficulties take place when finding a solution using the Heitler-Ma method [11]. In this context, it should be noted that in [6] the reduced system of Schrëdinger equations was used in modeling the excitation spectrum of the selective RF of the particles under consideration, but its spectrum was determined on the basis of the Heitler-Ma expressions that could be obtained without the under-consideration solutions $b_k(t)$.

Let us limit the consideration of $b_k(t)$ to the time t, when it can be assumed that the process of converting an irradiation photon into a RF photon has ended [11], the population of the initial state of the composite system is equal to zero and populations of excited collective states of particles are equal to zero. Let us determine the solutions to the above system of Schrëdinger equations for the states $|12\sigma\rangle$, $|12\nu\rangle$ at the above-specified t (which will be denoted as $t = \infty$), using the obtained expressions for $G_k(E)$ and the following relationships:

$$\lim_{t\to\infty}\xi(E - E_{12\sigma})\exp[i(E_{12\sigma} - E)t/\hbar] = -2\pi i\delta(E - E_{12\sigma}),$$

$$\int_{-\infty}^{+\infty} f(E)\cdot\delta(E - E_{12\sigma})dE = f(E_{12\sigma}).$$

This yields:

$$b_{12\sigma}(\bar\omega_\sigma, t = \infty) = i(2\pi)^{-1}$$
$$\lim_{t\to\infty}\int_{-\infty}^{+\infty} G_{12\sigma}(E)\cdot\exp[i(E_{12\sigma} - E)t/\hbar]dE$$
$$= -i\hbar^{-1}\Omega[V^{40}_{1\sigma}(\bar\omega_\sigma + 2\Delta + \gamma_3)]/F_\sigma(\bar\omega_\sigma),$$
$$b_{12\nu}(\bar\omega_\nu, t = \infty) = i(2\pi)^{-1}$$
$$\lim_{t\to\infty}\int_{-\infty}^{+\infty} G_{12\nu}(E)\cdot\exp[i(E_{12\nu} - E)t/\hbar]dE$$
$$= -i\hbar^{-1}\Omega V^{30}_{2\nu}w/F_\nu(\bar\omega_\nu),$$
$$F_\sigma(\bar\omega_\sigma) = (\bar\omega_\sigma - \bar\omega_I)[(\bar\omega_\sigma + 2\Delta + i\gamma_3)$$
$$\times(\bar\omega_\sigma + i\gamma 4) - w^2] - \Omega^2(\bar\omega_\sigma + 2\Delta + i\gamma_3),$$

and the function $F_\nu(\bar\omega_\nu)$ differs from $F_\sigma(\bar\omega_\sigma)$ by the $\bar\omega_\sigma$ replaced with $\bar\omega_\nu$. Using the definitions introduced above, let us write expressions for the population of the ground state of particles as follows:

$$\rho_{11}(t = \infty) = \hbar^{-2}\Omega^2\sum_\sigma |V^{40}_{1\sigma}|^2[(\bar\omega_\sigma + 2\Delta)^2$$
$$+ \gamma_3^2]/|F_\sigma(\bar\omega_\sigma)|^2,$$
$$\rho_{22}(t = \infty) = \hbar^{-2}\Omega^2\sum_\nu |V^{30}_{2\nu}|^2 w^2/|F_\nu(\bar\omega_\nu)|^2,$$

where

$$|F_\sigma(\bar\omega)|^2 = F^2_{\sigma 1}(\bar\omega_\sigma) + F^2_{\sigma 2}(\bar\omega_\sigma),$$
$$F_{\sigma 1}(\bar\omega_\sigma) = \mathrm{Re}F_\sigma(\bar\omega_\sigma) = (\bar\omega_\sigma - \bar\omega_L)$$
$$\times[(\bar\omega_\sigma + 2\Delta)\bar\omega_\sigma - \gamma_3\gamma_4 - w^2] - \Omega^2(\bar\omega_\sigma + 2\Delta),$$
$$F_{\sigma 2}(\bar\omega_\sigma) = \mathrm{Im}F_\sigma(\bar\omega_\sigma) = (\bar\omega_\sigma - \bar\omega_L)$$
$$\times[(\bar\omega_\sigma + 2\Delta)\gamma_4 + \bar\omega_\sigma\gamma_3] - \Omega^2\gamma_3,$$

and $|F_\nu(\bar\omega_\nu)|^2$ differs from $|F_\sigma(\bar\omega_\sigma)|^2$ by the $\bar\omega_\sigma$ replaced with $\bar\omega_\nu$.

By using the following equality:

$$\hbar^{-2}|V^{40}_{1\sigma}(\omega_\sigma = \omega_{41})|^2 = L^{-3}2\pi\hbar^{-1}\omega_{41}(d_\sigma)_{41}$$

(see the above note made regarding the determination of γ_4), after replacing the summation over indices σ with integration over all directions of photon σ propagation taking into account two states of its polarization [9], the following is obtained:

$$\hbar^{-2}\sum_\sigma |V^{40}_{1\sigma}(\omega_\sigma = \omega_{41})|^2 = \pi^{-1}\gamma_4.$$

Taking into account the above, we pass on from the summation of $\rho_{11}(t = \infty)$ over indices σ to the integration over $d\bar\omega_\sigma$, from the summation of $\rho_{22}(t = \infty)$ over indices ν to the integration over $d\bar\omega_\nu$ and get the following:

$$\rho_{11}(t = \infty) = \pi^{-1}\gamma_4\Omega^2$$
$$\times\int_{-\infty}^{+\infty} d\bar\omega_\sigma[(\bar\omega_\sigma + 2\Delta)^2 + \gamma_3^2])/|F_\sigma(\bar\omega_\sigma)|^2,$$

$$\rho_{22}(t = \infty) = \pi^{-1}\gamma_3\Omega^2\int_{-\infty}^{+\infty} d\bar\omega_\nu w^2/|F_\nu(\bar\omega_\nu)|^2.$$

For example, by determining the numerical values of these integrals for $\gamma_3 = 3$, $w = 5$, $\Delta = 10$ (hereinafter, unless otherwise noted, all particle parameters are given in γ_4) for the case of irradiation of molecule A with a pulse of duration of $T = 100\gamma_4^{-1}$ with a carrier frequency of $\bar\omega_L = 0$ and such an intensity that $\Omega = 0.25\gamma_4$, we get $\rho_{11}(\omega) = 0.8450$, $\rho_{22}(\omega) = 0.1550$, which reflects the preservation of the adopted normalization of amplitudes and coincides with the result of the numerical solving the system of equations given below for the dynamics of all states of particles with the corresponding selective RF.

Let us consider the obtaining expressions for the population and coherence of particle states and the relationships between them and their time derivatives for all time values of the selective RF process based on the relationship between the amplitudes of the states of a composite system and their time derivatives, which is determined by the above system of Schrëdinger equations.

Modeling the dynamics of population of particle states

Similar to what was adopted above for the consideration of dynamics of SF states, let us denote as $\rho_{11}(t)$ the probability of detecting particle A in the state $|1\rangle$, which is resulted from the $|4\rangle \to |1\rangle$ transition. Following the general principles of the description of selective fluorescence [4–6], $\rho_{11}(t)$ will be understood as the matrix element of the density operator of molecule A, obtained by reducing the $\rho_{12\sigma 12\sigma}(t)$ element of the density matrix of the composite system over σ photon states if $b_{12}\sigma(t)$ is a solution to the above-mentioned system of Schrëdinger equations. That is, the following definition will be used: $\rho_{11}(t) \equiv \sum_\sigma |b_{12\sigma}(t)|^2$. In other words, the $\rho_{11}(t)$ will be considered as a partial spur of the full density matrix, reflecting the „individual trajectory“ of states of the composite system in the subspace $|\sigma\rangle$ when a RF photon is absorbed.

Using this definition of $\rho_{11}(t)$ and the equation for $\dot{b}_{1\sigma}(t)$ in the above-mentioned system of Schrëdinger equations, we get

$$\dot{\rho}_{11}(t) = \frac{d}{dt}\sum_\sigma |b_{12\sigma}(t)|^2 = \hbar^{-2}\sum_\sigma |V_{1\sigma}^{40}|^2 b_{420}(t)$$

$$\times \int_0^t b_{420}^*(\tau)\exp[i\bar{\omega}_\sigma(t-\tau)]d\tau + c.c.$$

Applying the usual transition from summation over photon indices σ to integration over $d\bar{\omega}_\sigma$ and integration over the direction of photon propagation σ, taking into account two states of its polarization, we obtain, respectively, the term $\delta(t-\tau)$ and the term $\gamma_4 = 2\omega_{41}^3 d_{41}^2/3\hbar c^3$ (radiative decay constant of the excited state of molecule A [11], $\gamma_4 \ll \omega_{41}$):

$$\dot{\rho}_{11}(t) = \gamma_4 b_{420}(t)\int_0^t b_{420}^*(\tau)\delta(t-\tau)d\tau + c.c.$$

As follows from what is stated in [11], the amplitude $b_{420}(t \le 0) = 0$ is a regular function of time for $t = 0$. Also bearing in mind the considerations presented in [11] regarding the choice of limits in the integral notation of the function $\delta(t)$ for determining the form of functions regular for $t = 0$, the lower limit of integration over a variable τ will be replaced with $-\infty$, and the upper limit will be replaced with $+\infty$. Then, with the usual notation of

$$\int_{-\infty}^{+\infty} b_{420}^*(\tau)\delta(t-\tau)d\tau = b_{420}^*(t),$$

we get the $\dot{\rho}_{11}(t) = 2\gamma_4\rho_{44}(t)$ equation, which coincides in form with the above equation for the dynamics of particle states under selective SF.

Similarly, using the definition $\sum_\nu |b_{12\nu}(t)|^2 \equiv \rho_{22}(t)$ and the equation for $\dot{b}_{12\nu}(t)$, we derive

$$\frac{d}{dt}\sum_\nu |b_{12\nu}(t)|^2 \equiv \dot{\rho}_{22}(t) = \hbar^{-2}\sum_\nu |V_{2\nu}^{30}|^2 b_{130}(t)$$

$$\times \int_0^t b_{130}^*(\tau)\exp[i\bar{\omega}_\nu(t-\tau)]d\tau + c.c.$$

$$= 2\gamma_3|b_{130}|^2 \equiv 2\gamma_3\rho_{33}(t),$$

where

$$\gamma_3 = \pi\hbar^{-1}\sum_\nu |V_{2\nu}^{30}|^2\delta(E_{130} - E_{12\nu}) = 2\omega_{32}^3 d_{32}^2/3\hbar c^3$$

is radiative decay constant of the excited state of the particle, i.e. we have the following equation: $\dot{\rho}_{22}(t) = 2\gamma_3\rho_{33}(t)$.

It should be noted that $\dot{\rho}_{11}(t)$ and $\dot{\rho}_{22}(t)$ functions determine the intensity of emission of σ photons by particle A and ν photons by particle B, respectively, during the spectroscopic transitions mentioned above. Proportionality of the $\dot{\rho}_{11}(t)$ function to the population $|b_{420}(t)|^2 \equiv \rho_{44}(t)$ of the emitting state of particle A (and proportionality of the $\dot{\rho}_{22}(t)$ function to the population $|b_{130}(t)|^2 \equiv \rho_{33}(t)$ of the emitting state of particle B, respectively) is consistent with Beer's fundamental law [10]. Therefore, we postulate that the resulting equations $\dot{\rho}_{11}(t) = 2\gamma_4\rho_{44}(t)$ and $\dot{\rho}_{22}(t) = 2\gamma_3\rho_{33}(t)$ (without specifying the degree of rigor of their derivation) should be included in the desired system of equations that determine the population dynamics of the states of the particles under consideration under selective RF. As can be concluded from the form of the system of equations given in [4] for the population dynamics of particle states under selective RF, the desired system of corresponding equations under selective RF should also contain an equation of the following form:

$$\dot{\rho}_{33}(t) = -2\gamma_3\rho_{33}(t) - 2w\mathrm{Im}[\rho_{(13,42)}(t)\exp(i2\Delta t)].$$

It will reflect the decay of the excited basis state $|30\rangle$ and the interaction of particles. The values of $\rho_{33}(t)$ and $\rho_{(13,42)}(t)$ functions at $\Omega = 0$ should coincide with the values of the corresponding functions in [4] (see below the explanation of the $\rho_{(13,42)}(t)$ notation). And the right part of the equation for $\dot{\rho}_{44}(t)$ of the desired system of equations should include the $2w\mathrm{Im}[\rho_{13,42}(t)\exp(i2\Delta t)]$ term, the — $2\gamma_4\rho_{44}(t)$ term, as well as the term that determines the increase in population of the $|420\rangle$ state, which occurs due to a decrease in the population of the initial state $|12\lambda\rangle$, i.e. function $|b_{12\lambda}(t)|^2$, taken equal to 1 for $t = 0$. We will determine this function from the condition of preserving the normalization of the population of all basis states of the general system used, taking into account a separate consideration of the population of the ground state of each particle (selective consideration of subassemblies of individual particles according to [3]). Then, by denoting $|b_{12,\lambda}(t)|^2 \equiv \rho_{(12,12)}(t)$, we get

$$\rho_{(12,12)}(t) = 1 - \rho_{11}(t) - \rho_{22}(t) - \rho_{33}(t) - \rho_{44}(t),$$

and a conclusion can be made that the above-mentioned decrease in the population of the initial basis state $|12\lambda\rangle$ at RF in the desired system of equations should be described by such an equation for $\dot{\rho}_{(12,12)}(t)$, the right part of which is the term taken with the opposite sign in the equation for $\dot{\rho}_{44}(t)$, describing the corresponding increase in the value of the $\rho_{44}(t)$ function. This term will be selected based on a comparison between the expected desired system of equations for the population of particle states under selective RF and the corresponding OBE system (with the refinement of the adopted notation of $\rho_{33}(t)$ and $\rho_{44}(t)$ presented below). Thus, the form of all self-consistent equations that describe the $\rho_{11}(t)$, $\rho_{22}(t)$, $\rho_{33}(t)$, $\rho_{44}(t)$, and $\rho_{(12,12)}(t)$ functions will be determined.

It seems natural to assume that the desired system, which fully describes the dynamics of particle states under the selective RF under consideration, in addition to equations for the population dynamics of particle states, should contain equations that determine the dynamics of off-diagonal elements of the density matrix of the composite system: $b_{420}b^*_{130}$, $b_{420}b^*_{120}$, and $b_{120}b^*_{130}$. In this case, the set of equations for $\dot{\rho}_{11}(t)$, $\dot{\rho}_{22}(t)$, and $\dot{\rho}_{(12,12)}(t)$ should differ significantly from the set of equations describing the population dynamics of the $|12\rangle$, $|13\rangle$, $|42\rangle$ basic collective states of particles in the OBE system for the RF under consideration. However, there are grounds to believe that the form of the equations that determine the dynamics of the mentioned off-diagonal elements of the density matrix of the composite system is similar to the form of the equations for the dynamics of corresponding coherences in the OBE system. To compare the desired system of equations with the OBE system, let us present the OBE system for the dynamics of particle states when particle A is exposed to a rectangular pulse of monochromatic light with the following electric field strength:

$$E(t) = 2e_0 \sin\omega_L \Theta(t)\Theta(T-t) \equiv e(t)\sin\omega_L t$$

($\Theta(\tau) = 1$ at $\tau \geq 0$ and $\Theta(\tau) = 0$ at $\tau < 0$), $|\bar{\omega}_L| \ll \omega_{41}$, $\hbar^{-1}e_0\langle 12|d|42\rangle \equiv \Omega$ is Rabi frequency, d is dipole moment operator of particle A. With the notation $V_4^1 = -i\hbar\Omega\exp(-i\omega_L t)$, in the rotating wave approximation (equivalent to the resonant approximation used above), the OBE system can be written in the following form [9]:

$$\begin{aligned}
\dot{\rho}_{12,12}(t) &= 2\gamma_4\rho_{42,42}(t) + 2\gamma_3\rho_{13,13}(t) \\
&\quad + 2e(t)\Omega\mathrm{Re}[\rho_{42,12}(t)\exp(i\bar{\omega}_L t)], \\
\dot{\rho}_{13,13}(t) &= -2\gamma_3\rho_{13,13}(t) - 2w\mathrm{Im}[\rho_{13,42}(t)\exp(i2\Delta t)], \\
\dot{\rho}_{42,42}(t) &= -2\gamma_4\rho_{42,42}(t) - 2e(t)\Omega\mathrm{Re} \\
&\quad \times [\rho_{42,12}(t)\exp(i\bar{\omega}_L t)] + 2w\mathrm{Im}[\rho_{13}\rho_{42}(t)\exp(i2\Delta t)], \\
\dot{\rho}_{42,13}(t) &= -(\gamma_3 - \gamma_4 + \gamma_{43})\rho_{42,13}(t) - e(t)\Omega\exp(-i\bar{\omega}_L t) \\
&\quad \times \rho_{12,13}(t) + iw[\rho_{42,42}(t) - \rho_{13,13}(t)]\exp(i2\Delta t), \\
\dot{\rho}_{42,12}(t) &= -(\gamma_4 + \gamma_{41})\rho_{42,12}(t) - 2e(t)\Omega\exp(-i\bar{\omega}_L t) \\
&\quad \times [\rho_{12,12}(t) - \rho_{42,42}(t)] - iw\rho_{13,12}(t)\exp(i2\Delta t), \\
\dot{\rho}_{12,13}(t) &= -(\gamma_3 + \gamma_{32})\rho_{12,13}(t) + e(t)\Omega\exp(i\bar{\omega}_L t)\rho_{42,13}(t) \\
&\quad + iw\rho_{12,42}\exp(i2\Delta t).
\end{aligned}$$

Here the elements $\rho_{m,n} \equiv \langle m|\rho|n\rangle$, m, and n are indices of the basic collective states of particles. Phenomenological constants γ_4 and γ_3 characterize the rate of „longitudinal" radiative decay of the excited state of molecules A and B and decay induced by inelastic collisions with molecules of the environment of the pair under consideration. Constants γ_{41}, γ_{32} and γ_{43} characterize the rate of elastic-collision-induced „transverse" relaxation of off-diagonal elements of the density matrix $\rho_{42,12}$, $\rho_{12,13}$, and $\rho_{42,13}$, respectively [9].

Bearing in mind the above OBE system, let us take into account the assumption made regarding the form of the desired system of equations for the dynamics of selective RF. Then, by denoting $b_{420}b^*_{130}$, $b_{420}b^*_{120}$, and $b_{120}b^*_{130}$ as $\rho_{(42,13)}(t)$, $\rho_{(42,12)}(t)$, and $\rho_{12,13}(t)$, respectively, assuming that $\Omega = \Omega_\lambda$, and considering the damping constants appearing in the OBE system and in the desired system of equations to be equal, we get the following:

$$\dot{\rho}_{(12,12)}(t) = 2e(t)\Omega\mathrm{Re}[\rho_{(42,12)}(t)\exp(i\bar{\omega}_L t)],$$

$$\begin{aligned}
\dot{\rho}_{11}(t) &= 2\gamma_4\rho_{44}(t), \\
\dot{\rho}_{22}(t) &= 2\gamma_3\rho_{33}(t), \\
\dot{\rho}_{33}(t) &= -2\gamma_3\rho_{33}(t) - 2w\mathrm{Im}[\rho_{(13,42)}(t)\exp(i2\Delta t)], \\
\dot{\rho}_{44}(t) &= -2\gamma_4\rho_{44}(t) - 2e(t)\Omega\mathrm{Re}[\rho_{(42,12)}(t)\exp(i\bar{\omega}_L t)] \\
&\quad + 2w\mathrm{Im}[\rho_{(13,42)}(t)\exp(i2\Delta t)], \\
\dot{\rho}_{(42,13)}(t) &= -(\gamma_3 + \gamma_4 + \gamma_{43})\rho_{(42,13)}(t) \\
&\quad - e(t)\Omega\exp(-i\bar{\omega}_L t)\rho_{(12,13)}(t) \\
&\quad + iw[\rho_{(44)}(t) - \rho_{33}(t)]\exp(i2\Delta t) \\
\dot{\rho}_{(42,12)}(t) &= -(\gamma_4 + \gamma_{41})\rho_{(42,12)}(t) - e(t)\Omega\exp(-i\bar{\omega}_L t) \\
&\quad \times \rho_{(12,12)}(t) - \rho_{(44)}(t)] \\
&\quad - iw\rho_{(13,12)}(t)\exp(i2\Delta t),
\end{aligned}$$

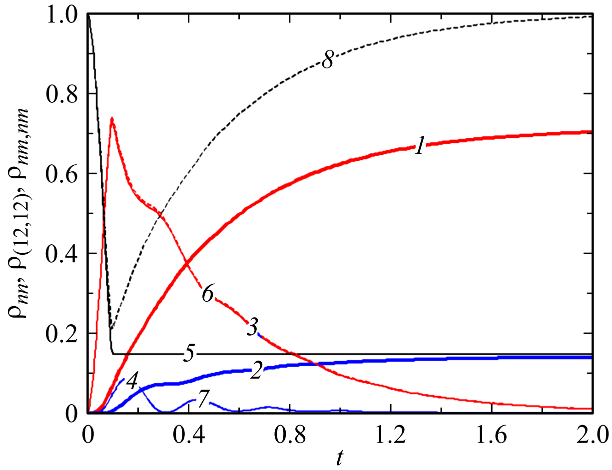

Figure 2. An example of the population dynamics of particle states under irradiation with a short pulse (see the text).

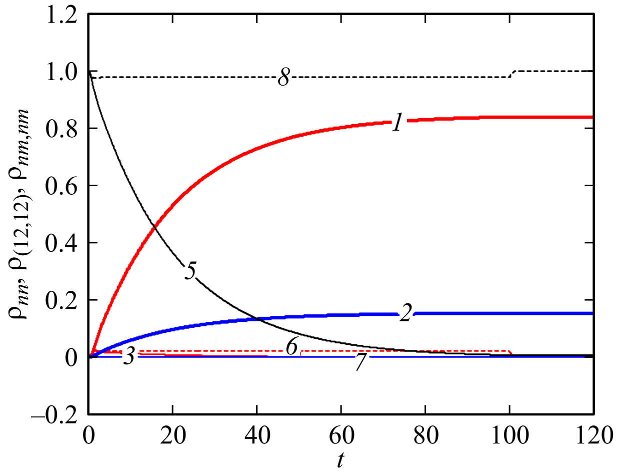

Figure 3. An example of the population dynamics of particle states under irradiation with a long pulse (see the text).

$$\dot{\rho}_{(12,13)}(t) = -(\gamma_3 + \gamma_{32})\rho_{(12,13)}(t)$$
$$+ e(t)\Omega \exp(i\bar{\omega}_L t)\rho_{(42,13)}(t) + iw\rho_{(12,42)} \exp(i2\Delta t).$$

This system of equations for $\Omega = 0$ and $\rho_{(12,12)}(t = 0) = 0$ coincides with the system of equations for the dynamics of selective SF given in [4] at the initial excitation of particle A by an ultrashort light pulse. So the resulting equations are an extension of the corresponding equations [4] to the case of irradiation of a particle with a long light pulse.

It is worth reminding that the probability $|b_{420}(t)|^2$ is denoted as $\rho_{44}(t)$, and the probability $|b_{130}(t)|^2$ is denoted as $\rho_{33}(t)$. This designation emphasizes their difference from the matrix elements $\rho_{42,42}(t)$ and $\rho_{13,13}(t)$ in the above OBE system, which does not reflect the influence of measuring the states of the irradiation field on the population of particle states. For example, as will be seen from the figures below, for times $\gamma_4^{-1} < t < T \gg \gamma_{4^{-1}}$ the $\rho_{44}(t)$ and $\rho_{42,42(t)}$ dependencies (functions) differ significantly from each other, as well as the $\rho_{33}(t)$ and $\rho_{13,13}(t)$ dependencies (cf. curves *3* and *6* and curves *4* and *7*, respectively, in Fig. 2–5).

The resulted system of equations for $\Omega = 0$ and $\rho_{(12,12)}(t = 0) = 0$ coincides with the system of equations for the dynamics of the selective SF presented in [4].

The difference between the yielded system of equations and the above OBE system is as follows. First, the OBE system contains one equation for the element $\dot{\rho}_{12,12}(t)$, and the yielded system contains three equations: for $\overline{\rho}_{(12,12)}(t)$, for $\dot{\rho}_{11}(t)$, and for $\dot{\rho}_{22}(t)$. Second, the equation for $\dot{\rho}_{(42,12)}(t)$ of the yielded system includes the element $\rho_{(12,12)}(t)$, and the corresponding equation of the OBE system includes the element $\rho_{12,12}(t)$. These elements differ little at $\gamma_4 t \ll 1$. Their difference can also be neglected for $t > T$. However, for the times $\gamma_4^{-1} < t < T >> \gamma_4^{-1}$ the above-mentioned elements differ significantly. This will be illustrated by the given figures (cf. curves *5* and *8* in Fig. 3). At the same time, it should be noted that the equations of the yielded

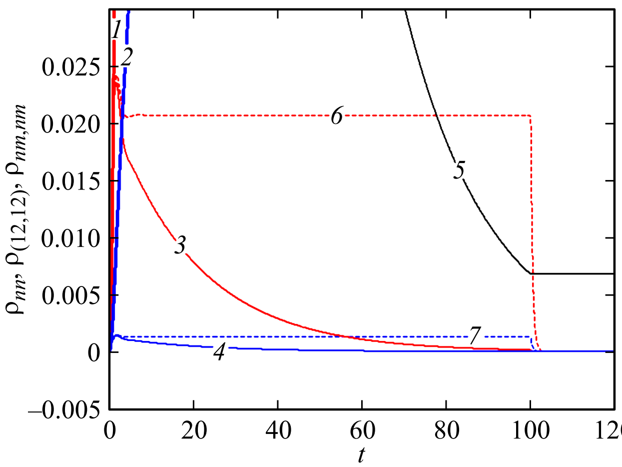

Figure 4. Zoomed-in part of Fig. 3 (see the text).

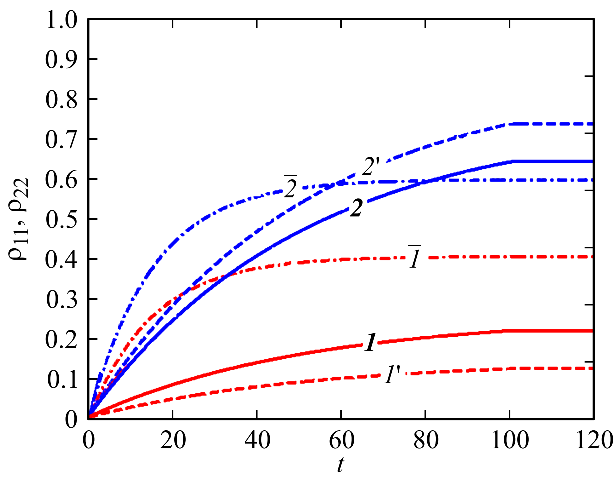

Figure 5. An example of the effect of the coherence of collective excited states of particles on the population dynamics of the ground state of each particle (see the text).

system relate the element $\rho_{(42,12)}(t)$ to $\rho_{44}(t)$, the element $\rho_{44}(t)$ to $\dot{\rho}_{11}(t)$ and, therefore, to $\dot{\rho}_{22}(t)$.

It is worth noting that, in accordance with the Introduction, the performed modeling of the RF dynamics was based on the idea of the transformation of one of the irradiation photons by particles. Thus, one of the eight self-consistent equations of the resulting system describes the dynamics of the population of the collective initial state of the composite system and the irradiation field, the other five equations describe the dynamics of the population and coherence of the collective states of particles and the state of the irradiation field without one of its photons, and each of the two remaining equations describes the population dynamics of the ground state of one of the particles, reflecting the transformation of an irradiation photon into an RF photon by this particle.

Examples and illustration of numerical solutions

To consider the characteristic features of the population dynamics of states of two-level particles predicted by the performed modeling and compare them with the results of similar modeling using the OBE formalism, we use the results of numerical solutions to the yielded system of equations and the OBE system, for simplicity considering strictly resonant irradiation ($\bar{\omega}_L = 0$). Fig. 2–4 show time dependences of the population of particle states found as a result of numerical solving of the yielded system and the corresponding OBE system. The abscissa axis shows time t in γ_4^{-1} units, and the ordinate axis shows the values of $\rho_{(12,12)}(t)$ functions, $\rho_{nn}(t)$ functions at $n = 1-4$, and $\rho_{nm,nm}(t)$ functions at $nm = 12, 13, 42$. Solid curves show $\rho_{11}(t)$ — red thickened curves (*1*), $\rho_{(22)}(t)$ — blue thickened curves (*2*), $\rho_{44}(t)$ — red curves (*3*), $\rho_{33}(t)$ — blue curves (*4*), $\rho_{(12,12)}(t)$ — black curves (*5*). Dashed curves show time dependences of the population of particle states according to the OBE system at the same values of particle parameters and irradiation: $\rho_{(42,42)}(t)$ — red dashed curves (*6*), $\rho_{13,13}(t)$ — blue dashed curves (*7*), $\rho_{12,12}(t)$ — black dashed curves (*8*).

The comparison with results of [4] is presented in Fig. 2, which illustrates the population dynamics of particle states with parameters $\gamma_3 = 3$, $w = 5$, $\Delta = 10$ (unless otherwise noted, all particle parameters are given in γ_4 units) upon irradiation of molecule A with a short pulse with a duration of $T = 0.1\gamma_4^{-1}$ and such an intensity that $\Omega = 12\gamma_4$. As can be seen from this figure, with the above-mentioned short irradiation pulse, the population dynamics of the collective excited states of particles in the OBE and in the yielded system are almost the same. After the end of the irradiation pulse, the population of the collective ground state of a pair of molecules, according to the OBE formalism, increases, tending to 1, and the populations of the ground state of each particle increase to their limiting value. In this case, the sum of the terms $\rho_{11}(\infty) \cong 0.7$, $\rho_{22}(\infty) \cong 0.15$, $\rho_{(12,12)} \cong 0.15$ is equal to 1. Hereinafter, when functions of time are considered for $t \gg \gamma_4^{-1}$, they are written as functions for $t = \infty$. The ratio of $\rho_{22}(\infty)/\rho_{11}(\infty) \cong 0.2$ corresponds to the preferential emission of an RF photon by particle A.

Fig. 3 and 4 illustrate the population dynamics of particle states with the same parameters $\gamma_3 = 3$, $w = 5$, $\Delta = 10$ when particle A is excited by a pulse duration $T = 100\gamma_4^{-1}$ and such an intensity that $\Omega = 2.25\gamma_4$. Fig. 3 illustrates the smooth decrease of $\rho_{(12,12)}(t)$, $\rho_{33}(t)$, and $\rho_{44}(t)$ functions down to $\cong 0$ with $t \to \infty$ and the corresponding smooth increase of $\rho_{11}(t)$ and $\rho_{22}(t)$, so that $\rho_{11}(\infty) + \rho_{22}(\infty) \cong 1$. In this case, the values of $\rho_{11}(\infty)$ and $\rho_{22}(\infty)$ are consistent with the values of $\rho_{11}(\infty) = 0.8450$, $\rho_{22}(\infty) = 0.1550$ given in the previous part of the article, which are calculated on the basis of the exact solution to the above system of Schrëdinger equations for the RF at $t = \infty$. The dashed lines reflect the stationary state of populations $\rho_{12,12}(t)$, $\rho_{42,42}(t)$, and $\rho_{13,13}(t)$ at $\gamma_4^{-1} < t < T$. Fig. 4 shows some of the mentioned curves zoomed-in.

Fig. 3 and 4 illustrate the consequences of the difference in the dynamics of the population of particle states described by the yielded system and the OBE system. Thus, all solid curves reflect the dynamics of the transformation of one of the irradiation photons into a RF photon: a decrease in the population of the initial state of the composite system (curve *5*), a decrease in the population and coherence of excited collective states of particles (curves *3* and *4*), an increase in the population of the final states of each particle (curves *1* and *2*), and the dashed straight lines reflect the stationary transformation of the classically described „reabsorbed“ irradiation field by particles.

Fig. 5 illustrates an example of the above-mentioned effect on $\rho_{nm}(t)$ of the yielded system of equations from its coherences for particles with parameters $\gamma_3 = 3$, $w = 20$, $\Delta = 10$ and irradiation with $T = 100\gamma_4^{-1}$, at which $\Omega = \gamma_4$. In the figure, the ordinate axis shows the values of $\rho_{11}(t)$ and $\rho_{22}(t)$ functions. These functions, calculated without taking into account the transverse relaxation constants of all coherences in the yielded system (as well as the values of the functions shown in the previous figures), are shown by solid curves *1* and *2*, respectively. The dashed curves *1′* and *2′* represent $\rho_{11}(t)$ and $\rho_{22}(t)$ functions, respectively, calculated for particles with the above parameters with additional consideration of one constant: $\gamma_{43} = 10\gamma_4$. The dash-dot curves, $\bar{1}$ and $\bar{2}$, represent $\rho_{11}(t)$ and $\rho_{22}(t)$ functions, respectively, calculated for molecules with the above parameter values with additional consideration of $\gamma_{41} = \gamma_{32} = 10$ constants. The same figure can be considered as an illustration of taking into account the effect of elastic collisions (reflected by an increase in the rate constants of transverse relaxation of the elements $\rho_{(42,12)}$, $\rho_{(12,13)}$, and $\rho_{(42,13)}$) on the dynamics of selective RF.

By comparing the solid curves *1* and *2* with the dashed curves *1′* and *2′*, respectively, it should be concluded that the increase in coherence $\rho_{(42,13)}$ resulted from taking into account the constant $\gamma_{43} = 10\gamma_4$ (reflecting elastic collisions of particles with each other) increases the efficiency of RF photon emission by particle B. In this case, $\rho_{22}(t)$ increases and $\rho_{11}(t)$ decreases by the same amount, without changing the dynamics of $\rho_{(12,12)}$. Comparing the curves *1* and *2* with $\bar{1}$ and $\bar{2}$, respectively, it should be concluded that elastic collisions of particles with particles of the environment (taking into account $\gamma_{41} = \gamma_{32} = 10\gamma_4$) increase the efficiency of RF photon emission by particle A with a significant increase in $\rho_{11}(t)$ and with a slight decrease in $\rho_{22}(t)$, starting from $t > 80\gamma_4^{-1}$.

Concluding note

It is shown that the dynamics of the selective RF of two interacting nanoparticles, described by the yielded system of equations, differs significantly from the dynamics of the RF of these particles, which is described by the system of equations of the corresponding density matrix of their collective states. It seems useful to keep this in mind when using RF as a method for determining the parameters characterizing the dynamics of interacting nanoparticles irradiated by light when considering the possibility of their use as materials for various optoelectronic devices.

Conflict of interest

The author declares that he has no conflict of interest.

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