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Mathematical modeling of resonance fluorescence spectra of two two-level interacting nanoparticles

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The mathematical modeling of the resonance fluorescence spectra associated with the dynamic interaction of two two-level nanoparticles is carried out. The cases of weak and strong pumping of a single particle by a long pulse of monochromatic light with a carrier frequency equal to or close to the frequency of its own transition are considered. Expressions for the contours of the spectra of collective and two components of selective resonant fluorescence are obtained — as a function of the frequency of the recorded photon of fluorescence — as a result of using solutions of the Schrodinger equation system for the amplitudes of states of a composite system of particles, a quantized irradiation field and a resonant fluorescence field. Based on the analysis of graphical images of the obtained functions, the characteristic features of the shape and position of the maxima of the contours of the spectra at different values of the parameters of the structure and interaction of particles and the irradiation field are determined.

Keywords: modeling, two-level nanoparticles, resonance fluorescence spectrum.

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Introduction

Mathematical modeling of the resonance fluorescence spectrum of two-level interacting nanoparticles (models of molecules, atoms, ions, and semiconductor quantum dots) irradiated with monochromatic light reflects the dependence of a number of characteristics of the RF frequency filtration field on the values of irradiation parameters and particle parameters. The results of calculations of this dependence can be used for comparison with the corresponding experimental data of the RF for obtaining information about the mutual arrangement of particles, the nature of their interaction with each other and with the radiation field. A comparison of the results of such calculations obtained during modeling using various variants of the light conversion mechanism by particles allows drawing conclusions regarding the confirmation of the adequacy of the accepted modeling.

Researchers quite often consider the dipole-dipole interaction of stationary atoms the distance between which is less than the wavelength of the irradiated light, equal to the wavelength of the atoms' proper transitions. The guideline for such modeling is usually the modeling of the RF spectrum of one two-level atom conducted in numerous studies. The expression for the RF spectrum of an atom is determined based on consideration of the Fourier component of the RF field correlation function, which is expressed in terms of the steady-state solution of the governing equation for the atomic density matrix (in the Markov, Born approximation), taking into account the decay of the atom excited state according to Lindblad. The dependence of the stationary population of an atom on the frequency of the recorded photon of fluorescence is also used as the definition of the RF spectrum. According to the results of these studies, it was found that the RF spectrum has the form of the one line in case of a low radiation intensity with the spectrum contour having the form of almost one line, and in case of a high radiation intensity, when the frequency of oscillation of the population levels of an atom is much greater than the decay rate constant of its excited state, the RF spectrum has the form of a combination of three contours („peaks"): central (with a maximum frequency equal to the frequency of irradiation) and two „lateral", the maximum frequency of one of them is greater and the other is less than the frequency of irradiation by the frequency of radiation. The width of the central peak is equal to half, and the width of the lateral peaks is equal to three quarters of the value of the decay constant. These conclusions are consistent with the data of the corresponding measurements of the RF spectrum of one atom, therefore, in modeling the RF spectrum of two atoms, the main provisions of the general theoretical approach noted above are used in modeling the RF spectrum of one atom, and the corresponding computational formalism takes into account the interaction of atoms, for example, dipole-dipole. The paper with the title „Collective atomic effects in resonance fluorescence: Dipole-dipole interaction" is one of the first of such studies [1].

The RF spectrum of a composite system of two identical two-level atoms interacting with each other and with a quantized field of monochromatic irradiation with a frequency equal to the frequency of natural transitions of stationary atoms, the distance between which does not exceed the wavelength of irradiation, is modeled in this study. The dynamic dipole-dipole interaction of atoms and the following collective states of the system were taken into account: 1) a state in which both atoms are in their ground states, and the irradiation field contains a certain, rather large number of photons, 2) two states, in each of which one of the atoms is in an excited state, the other — mainly, and the irradiation field contains one photon less initial, and 3) a state in which both atoms are excited, and the irradiation field contains two photons less than the initial one. This study determines such energy levels and corresponding eigenstates of the system which reflect the splitting of the degenerate excited energy levels of the system due to the dipole-dipole interaction of atoms with each other and with the irradiation field. These states are used as the basic states for the formulation of the control equation for the density matrix of the system, which takes into account the decay of excited states of atoms according to Lindblad. Approximations which are valid for one or another relative value of the atomic parameters and the radiation intensity were used for solving this equation. Various methods of solving the equation were chosen for cases when a clear separation of lateral contours of the spectrum from the central contour was expected in case of a particular force of interaction of atoms with separation of these contours from each other and when their significant overlap was expected. The obtained stationary solutions are applied for composing an expression for the time-average value of the correlation function of the dipole operator of atoms, used for determining the expression for the first-order correlation function of the field in the wave zone. The expression for the RF spectrum is obtained using the Fourier transform of this correlation function. As a result of the analysis of the obtained expression, it is concluded that the RF spectrum looks (in the general case) as two groups of contours that are symmetrically offset relative to the central contour on both sides with the peak frequency being equal to the frequency of irradiation of atoms in case of different values of the parameters of atoms, their interaction and radiation intensity. The displacement of the peaks of the lateral contours is greater than the frequency of the Rabi transitions in atoms and the greater the energy of the dipole-dipole interaction of atoms. When considering the case of irradiation with light of such intensity that the Rabi frequency is 14 in units of the spontaneous emission rate constant of each atom, Fig. 2 in [1] shows the images of example spectra at 11 different values of the parameter characterizing the strength of the dipole-dipole interaction between the atoms. It is noted in the abstract to this work that the significant difference between the RF spectrum of two atoms and the RF spectrum of one atom predicted by the simulation is inconsistent with the conclusions of previous study (reference is given in the literature list), which used the same general approach to modeling the RF spectrum of two atoms, but without taking into account their dipole-dipole interaction.

In subsequent years, a number of studies of various authors simulated the spectrum of single-photon collective resonance fluorescence from two interacting two-level atoms resonantly irradiated by monochromatic light. These simulations employed a classical description of the irradiation and accounted for both the coherent and incoherent parts of the dipole-dipole interaction between stationary atoms separated by a distance no greater than the wavelength of the driving field. The expression for the spectrum and the computational formalism were chosen based on the corresponding descriptions of modeling the RF spectrum of a single atom. One of the latest of such papers provides an image of the RF spectrum (Fig. 1, *b* in [2]) for the case of irradiation, when the Rabi frequency is 30 times higher than the decay rate constant of excited states of atoms, with a selected characteristic value of the dipole-dipole energy interactions of atoms. This spectrum looks like a central contour with a maximum at the irradiation frequency and three lateral contours symmetrically located on both sides of the central one. Two of these lateral circuits are located in the frequency range shifted from the irradiation frequency to the Rabi frequency, and the third lateral circuit is — at twice the Rabi frequency. All contours in the area of their maxima look like „peaks“, but overlap with each other at about half their height. The height of the maximum of the central peak slightly exceeds approximately the same height of the peaks in the region shifted by the Rabi frequency, and the height of the maximum of these peaks significantly exceeds the height of the peak shifted by twice the Rabi frequency.

A theoretical approach to modeling the collective RF spectrum of two atoms, which differs from the one used in Ref. [1,2], is applied in Ref. [3]. This paper considers the RF of a composite system of atoms irradiated with classically described monochromatic light and the RF photon field. The following states were distinguished among the collective states of this composite system: 1) a state in which the field contains an RF photon, and both atoms are in their ground states; 2) two states, in each of which one atom is in an excited state, the other — in the ground state, while the field contains an RF photon; 3) a state in which both atoms are excited, and the field contains an RF photon. A governing equation is compiled for the elements of the density matrix of such a composite system using these states as the basic ones. Radiative decay of the excited atomic states was introduced by reducing the system's density matrix over the zero-photon field states while accounting for the considered incoherent dipole-dipole interaction between the atoms. Using a computational formalism similar to the formalism used earlier in a number of works with this approach to modeling the RF spectrum of a single atom, stationary solutions of the control equation for the density matrix of the system are obtained. It was taken into account that a number of approximations were used that were valid for the selected relative values of the atomic parameters and their position relative to the direction and polarization of the irradiation. For example, the impact of non-diagonal elements of the density matrix on the diagonal ones is neglected and the colonization of the collective state level at which both atoms are excited is excluded. The expression for the spectrum was defined as the dependence of the sum of the stationary populations of the first three of the collective states of the system noted above on the frequency of the RF photon. The expression defined in this way for the RF spectrum is written in an analytical form. It is obtained if the following conditions are met: 1) precise resonance with intrinsic transitions of identical atoms, 2) neglect of atomic level shifts due to their dipole-dipole interaction, 3) the Rabi frequency is much higher than the radiative decay constant of the excited state of each atom. The form of the obtained RF photon frequency function is shown in the figure (Fig. 1 in [3]), which illustrates the authors' concluding remark that the spectrum contains wide side bands symmetrically arranged relative to the central band with a shift by the Rabi frequency and an additional, wider band with a small maximum. shifted by about twice the Rabi frequency. It was noted in the final part of this paper that such a prediction regarding the shape of the RF contour does not agree with predictions of the shape of the RF made in a number of different works for the same values of atomic parameters and irradiation that were obtained using several other general approaches (references to 5 such papers are given).

In addition to the differences in RF spectral patterns reported in [1–3], there are also varying conclusions about the general characteristics of these RF spectra across different studies that we would like to add. This applies, for example, to the following conclusions: 1) the difference between the RF spectrum of two atoms and the RF spectrum of one atom with low radiation intensity is more pronounced than with the difference with high radiation, 2) the asymmetric shift of the lateral contours relative to the central one, 3) the origin of the lateral contours, which are shifted on both sides of the central one by twice the frequency of radiation. In the absence of a comparison of the obtained spectra with the corresponding experimental data, which would clarify the origin of these inconsistencies, it can be assumed that these differences are attributable to differences in the approaches and (or) computational formalism used in the mentioned studies.

Bearing in mind the mentioned disagreement, it is of interest to have the results of modeling the RF spectrum of two atoms, which was performed using a theoretical approach to considering RF of two atoms, which differs from the approaches used in Ref. [1–3]. As such, we can take the approach used in modeling the RF excitation spectrum of two atoms in Ref. [4] and the dynamics of population and coherence of states of two atoms in case of RF in Ref. [5].

The goal of this study is the modeling of the RF spectrum of two atoms when applying the theoretical approach to the consideration of RF, which is used in Ref. [4,5]. Among the main differences between this modeling and the one used in the above studies, we would like to note the following: 1) using quantum consideration of the states of both the irradiation field and the scattered field, 2) composing an expression for the RF spectrum as a dependence of the separately considered population of the ground state of each atom on the frequency of the detected RF photon, attributing it to radiation from one atom or another („selective RF"), 3) determination of these populations as a result of a rigorous solution of the Schrodinger equation system for the amplitudes of the considered states of a composite system of atoms and a radiation field, 4) consideration of RF atoms with different frequencies of their own transitions under both strictly resonant and quasi-resonant irradiation.

The following parts of this paper provide a detailed description of all the details of performing the accepted modeling when applying the computational formalism of studies in Ref. [4,5], using terms such as „nanoparticle" (or just a particle) and „RF spectrum" used in Ref. [4,5]. Graphical images of the obtained RF photon frequency functions are presented, which illustrate examples of the dependence of the characteristic features of the shape and position of the maxima of the contours of the collective and selective RF spectra on the values of the parameters of the structure and interaction of particles and fields of resonant and quasi-resonant irradiation.

Model of a complete composite system

The Hamiltonian of a complete composite system of two „point" stationary two-level particles (particle A and particle B with their non-overlapping wave functions), which interact with each other and with a quantized radiation field, is written as

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

where H_f is the Hamiltonian of the radiation field; W is the energy operator of the dynamic interaction between particles, $H_A(H_B)$ is the Hamiltonian of particle $A(B)$, $V_A = -(\mathbf{ed}_A$ is the energy operator of the electrodipole ($\mathbf{d}_A$ is the operator of the dipole moment of particle A) interaction of particle A with the transverse components of the radiation field and a similar expression $V_B = -(\mathbf{ed}_B)$ for particle B. The orientation of the dipoles of the particle transitions and the direction of the irradiation can be such that it excites only the particle A. The distance between the particles can be less than or on the order of the wavelength of spontaneous fluorescence of an isolated particle and an isolated particle B.

By the operator W, which induces reversible transitions between particles in excited states, we shall mean the energy operator of dipole-dipole interaction between particles. This could be, for instance, the operator adopted in Ref. [1] to describe interactions between particles separated by distances comparable to the wavelength of their intrinsic transitions; or the operator used to describe interactions between closely spaced particles, such as: two alkaline-earth atoms whose fluorescence intensity beats were studied in Ref. [6]; or a pair of CdSe/ZnS quantum dots whose relative geometry was determined through analysis of their RF spectra in Ref. [7].

The eigenstates of a particle A with energies E_1 and E_4 are denoted by $|1\rangle$ and $|4\rangle$, and the eigenstates of a particle B with The energies E_2 and E_3 denote $|2\rangle$ and $|3\rangle$. For clarity, consider a pair of particles with $E_4 \geq E_3$, $E_3 \geq E_2$, $E_2 = E_1$ and use the notation $\hbar\omega_{nm} = (E_n - E_m)$, where $n, m = 4 - 1$, $n > m$, $\omega_{41} - \omega_{32} \equiv 2\Delta$, $0 \leq \Delta \ll \omega_{41}, \omega_{32}$.

The initial irradiation field contains an integer $(N > 1)$ of photons λ with a frequency ω_L such that $|\omega_L - \omega_{41}| \ll \omega_{41}$; λ is a set of indices characterizing the wave vector of a photon and the state of its polarization. The state of the field containing N of photons λ is denoted by $|\lambda\rangle$, and the state of the field containing $N - 1$ of photons λ is denoted by $|0\rangle$.

Let's use $|12\mu\rangle$ to denote such a state of a complete composite system when the radiation field contains $N - 1$ of photons λ and one photon μ $(\mu \neq \lambda)$ RF, and a pair of particles are in their „collective" ground state $|12\rangle$. For the sake of brevity, here and further, where this does not lead to misunderstanding, the index „0" is omitted in the notation $|12\mu\rangle$.

Assuming that the particles and the radiation field under consideration are located in the volume space L^3, let us denote the matrix element of the operator V_A by the states of the complete composite system $|12\lambda\rangle$ and $|420\rangle$ as

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

where $(d_\lambda)_{14}$ is the matrix element for states $|1\rangle$ and $|4\rangle$ (which is based on the real value) of the projection of the operator d on the direction of photon polarization λ, Ω is „work frequency". Let us use the following notation for the matrix element V_A, using the states $|12\mu\rangle$ and $|420\rangle$

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

where $(d_\mu)_{14}$ is the matrix element of the operator V_A according to the states $|1\rangle$ and $|4\rangle$ of projections of the operator d on the direction of photon polarization μ, ω_μ is the photon frequency μ. Let us use the following notation for the matrix element V_B, using the states $|12\mu\rangle$ and $|130\rangle$

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

where $(d_\mu)_{23}$ is the matrix element of the projection of the operator D onto the direction of photon polarization μ by states $|2\rangle$ and $|3\rangle$. For brevity, $V^{420}_{12\lambda}$ will be written as $V^{40}_{1\lambda}$, $V^{420}_{12\mu}$ will be written as $V^{40}_{1\mu}$ and $V^{130}_{12\mu}$ as $V^{30}_{2\mu}$.

Just as in Ref. [4,5], the Hilbert space of states $|0\sigma\rangle$ of the marked field $|0\rangle$ and the photon field σ, emitted in case of the RF by a particle A interacting with the particle B, and the states $|0\nu\rangle$ of the field $|0\rangle$ and the photon field ν emitted by the particle B in case of the considered RF. Let us omit the index „0" in the notation of these states, as well as in the notation of states $|\mu\rangle$. The corresponding matrix elements have the following form:

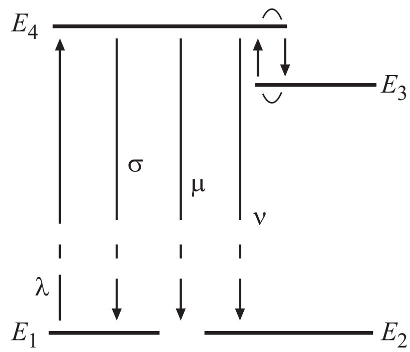

Figure 1. A diagram of the relative position of the energy levels of particles, indicating transitions when the corresponding states change.

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

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

The matrix elements of the operator W between the states $|420\rangle$ and $|130\rangle$ will be denoted as $\langle 42|W|13\rangle = \langle 13|W|42\rangle \equiv \hbar w$.

Fig. 1 schematically shows the relative position of the energy levels $E_1 \div E_4$ marked by horizontal line segments of the considered pair of particles. Circular curves indicate nonradiative, reversible transitions between particles in excited states due to the action of the operator W. Vertical lines with arrows represent radiation transitions in a complete composite system, which are accompanied by a change in the state of the radiation field and the field of collective or selective resonant fluorescence.

System of equations and its solutions

We will consider the modeling of the dynamics of the occupancy of states of a complete composite system based on the use of solutions to the Schrodinger equation system for the amplitudes $b_k(t)$ of the population of the eigenstates of the accepted Hamiltonian. This system has the following form in the accepted resonant approximation:

$$i\hbar\dot{b}_k(t) = \sum_l \langle k|(V_A + V_B + W)l\rangle|b_l(t)$$
$$\times \exp[i(E_k - E_l)t]/\hbar] + i\hbar\delta_{ki}\delta(t),$$

where i, k, l are the indices of the basic orthonormal states of the complete composite system: i are initial states $|12\lambda\rangle$, k and l are states $|420\rangle$, $|130\rangle$, $12\mu\rangle$; The energy of these states is denoted as E_i, E_k, E_l; $\delta(t)$ — Dirac function, δ_{ki} is the Kronecker symbol, $\delta_{ki} = 0$ for $k \neq i$ and $\delta_{ki} = 1$ for $k = i$. The time is counted from the moment $t = 0$.

Regarding the role of the heterogeneous member $i\hbar\delta_{ki}\delta(t)$ and the preservation of normalization $\sum_k |b_k(t)|^2 = 1$ — see, for example, § chapter 16 4 in Ref. [8].

The system of equations for the amplitudes of the basic states $|12\lambda\rangle$, $|420\rangle$, $|130\rangle$, $|12\mu\rangle$ under the initial condition $b_{12\lambda}(+0) = 1$ has the following form:

$$i\hbar\dot{b}_{12\lambda} = V^{40}_{1\lambda} b_{420}(t)\exp[i(E_{420} - E_{12\lambda})t/\hbar] + i\hbar\delta(t),$$

$$i\hbar\dot{b}_{420}(t) = V^{1\lambda}_{40} b_{12\lambda}\cdot\exp[i(E_{12\lambda} - E_{420})t/\hbar] + \sum_{\mu\neq\lambda} V^{1\mu}_{40} b_{12\mu}(t)\cdot\exp[i(E_{420} - E_{12}\mu)t/\hbar] + W^{13}_{42} b_{130}(t)\cdot\exp[i(E_{420} - E_{130})t/\hbar]$$

$$i\hbar\dot{b}_{130}(t) = \sum_{\mu\neq\lambda} V^{2\mu}_{30} b_{12\mu}(t)\cdot\exp[i(E_{130\lambda} - E_{12}\mu)t/\hbar] + W^{42}_{13} b_{420}(t)\cdot\exp[i(E_{130} - E_{420})t/\hbar],$$

$$i\hbar\dot{b}_{12\mu}(t) = V^{40}_{1\mu} b_{420}(t)\cdot\exp[i(E_{12\mu} - E_{420}t/\hbar] + V^{30}_{2\mu} b_{130}(t)\cdot\exp[i(E_{12\mu} - E_{130})t/\hbar].$$

With the accepted basis for describing the states of a complete composite system, the above system of equations reflects the consideration of the excitation by irradiation of only particles A. Such excitation can be realized by a certain choice of the appropriate configuration of the relative arrangement of particles, the direction of propagation and polarization of photons λ or by choosing the frequency ω_L, if the particles are such that $\Delta \gg \gamma_4$. It should be noted also that this system of equations describes the dynamics of the probability amplitudes of the reduced basic states of the complete composite system when one of the irradiation photons is converted into the corresponding RF photon, without taking into account various other particle states and the radiation field and, accordingly, other radiation processes of the RF photon. For example, such processes, about which the following remark is given on page 249, chapter 10 in Ref. [9]: „an atom can coherently interact with a field many times before spontaneously emitting a photon".

When using the Fourier representations of the amplitudes $b_k(t)$ and $\delta(t)$, the functions,

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

and

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

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

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

Turning to the solution of this system of equations, we use the following expressions:

$$i\hbar\gamma_4(E) = -\sum_\mu |V^{40}_{1\mu}|^2 \varsigma(E - E_{12\mu}),$$

$$\hbar\gamma_3(E) = -\sum_\mu |V^{30}_{2\mu}|^2 \varsigma(E - E_{12\mu}),$$

$$\hbar\tilde{\gamma}(E) = -\sum_\mu V^{40}_{1\mu} V^{2\mu}_{30} \varsigma(E - E_{12\mu}),$$

where $\varsigma(E) = P/E - i\pi\delta(E)$, P/E is the main value of the function $1/E$. Leaving in these sums only the terms proportional to $\delta(E)$, after performing the specified summation (see [8]), the resulting expressions will mean the constants $\gamma_4 = 2\omega^3_{41} d^2_{41}/3\hbar c^3$, $\gamma_3 = 2\omega^3_{32} d^2_{32}/3\hbar c^3$, $\tilde{\gamma} = 2\omega^3_{41} d_{41} d_{32}/3\hbar c^3$, which characterize, respectively, the rate of spontaneous radiative decay of the state $|4\rangle$ of an isolated particle A, the rate of spontaneous radiative decay of the state $|3\rangle$ of an isolated particle B and the rate of „joint" spontaneous radiation decay of these states. Such use of the above expressions when describing the RF of two interacting particles with the accepted condition $0 \leq \Delta \ll \omega_{41}, \omega_{32}$ seems to be as justified as the similar use of the constant $\gamma_4 = 2\omega^3_{41} d^2_{41}/3\hbar c^3$ instead of the expression

$$i\hbar\gamma_4(E) = -\sum_\mu |V^{40}_{1\mu}|^2 \varsigma(E - E_{12\mu}),$$

which is accepted when describing the RF corresponding to one isolated two-level particle in Ref. [8] — see the notes on page 230 and the text of the footnote on page 231.

Using the introduced notation, let us write the system of equations for $G_{ki}(E)$ (omitting the reference to the dependence of G_{ki} on E) as

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

$$(E - E_{420} + i\gamma_4)\cdot G_{420} = V^{1\lambda}_{40} G_{12\lambda} + \hbar(w - u\tilde{\gamma})G_{130},$$

$$(E - E_{130} + i\gamma_3)\cdot G_{130} = \hbar(w - i\tilde{\gamma})G_{420},$$

$$(E - E_{12\mu})\cdot G_{12\mu} V^{40}_{1\mu} G_{420} + V^{30}_{2\mu} G_{130}.$$

Solving this system of equations, we find

$$G_{12\mu}(E) = \left[V^{40}_{1\mu} V^{1\lambda}_{40} E_I + V^{30}_{2\mu} V^{1\lambda}_{40} \hbar(w - i\tilde{\gamma})\right] \times \varsigma(E - E_{12\mu}/F(E)),$$

where

$$F(E) = E_0\left[E_I E_{II} - \hbar^2(w^2 - \tilde{\gamma}^2) + 2i\hbar w\tilde{\gamma}\right] - \hbar^2\Omega^2 E_I,$$

$$E_0 = E - E_{12\lambda},$$

$$E_I = E - E_{130} + i\hbar\gamma_3,$$

$$E_{II} = E - E_{420} + i\hbar\gamma_4.$$

Using this expression for $G_{12\mu}(E)$ and the ratio

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

$$\int_{-\infty}^{\infty} f(E) \cdot \delta(E - E_{12\mu}) dE = f(E_{12\mu}),$$

for $t \gg 1/\gamma_4$ we obtain

$$b_{12\mu}(\overline{\omega}_\mu, t = \infty) = i(2\pi)^{-1} \lim_{t\to\infty} \int_{-\infty}^{\infty} G_{12\mu}(E)$$

$$\times \exp[i(E_{12\mu} - E)t/\hbar)]dE = -i\hbar^{-1}\Omega$$

$$\times \left[V_{1\mu}^{40}(\overline{\omega}_\mu + 2\Delta + i\gamma_3) + V_{2\mu}^{30}(w - i\tilde{\gamma})\right] / F_\mu(\overline{\omega}_\mu),$$

where $\overline{\omega}_\mu \equiv \omega_{\mu-\omega_{41}}$, $\overline{\omega}_L \equiv \omega_L - \omega_4$,

$$F_\mu(\overline{\omega}_\mu) = (\overline{\omega}_\mu - \overline{\omega}_L)\left[(\overline{\omega}_\mu + 2\Delta + i\gamma_3)(\overline{\omega}_\mu + i\gamma_4)\right.$$

$$\left. - w^2 + \tilde{\gamma}^2 + 2iw\tilde{\gamma}\right] - \Omega^2(\overline{\omega}_\mu + 2\Delta + i\gamma_3).$$

It should be noted that if $w = \gamma_3 = \Delta = \tilde{\gamma} = 0$, then $b_{12\mu}(\overline{\omega}_\mu, t = \infty)$ has the form of an expression for the amplitude of the RF state of one isolated particle A:

$$b_A^{RF}(\overline{\omega}_\mu, t = \infty) = \frac{V_{1\mu}^{40} V_{40}^{1\lambda}}{\hbar^2(\overline{\omega}_\mu - \overline{\omega}_L)(\overline{\omega}_\mu + i\gamma_4) - |V_{1\lambda}^{40}|^2}$$

$$= \frac{V_{1\mu}^{40} V_{40}^{1\lambda}}{\hbar^2(\overline{\omega}_\mu + i\gamma_4)\left[\overline{\omega}_\mu - \overline{\omega}_L + i\Gamma_\mu(\overline{\omega}_\mu)\right]},$$

where $\Gamma_\mu(\overline{\omega}_\mu) = i|V_{1\lambda}^{40}|^2/(\overline{\omega}_\mu + i\gamma_4)$.

The function $b_A^{RF}(\overline{\omega}_\mu, t = \infty)$ has the form of expression (20.10) given on page 231 in [8] after substituting expressions (20.6) and (20.7) given on page 230 into it.

The amplitude $b_{12}\mu(\overline{\omega}_\mu, t = \infty)$ is the sum of the terms, displaying „interfering alternatives“ (see pages 25, 26 in Ref. [10]) of the probability of photon emission from the RF through the „radiation channel $|4\rangle \to |1\rangle$“ particle A and „radiation channel $|3\rangle \to |2\rangle$“ particle B with the occupancy of the collective ground state of the particles. In practice, the detection of the RF field state can be such (see the remark on page 468 in Ref. [9], paragraph 21.1.3 of Chapter 21) that it „perceives only radiation at the junction $|4\rangle \to |1\rangle$ and ignores radiation at the junction $|3\rangle \to |2\rangle$, or vice versa, perceives radiation at the junction $|3\rangle \to |2\rangle$ and ignores radiation at the junction $|4\rangle \to |1\rangle$, choosing one or another „path of radiation“ RF photon by one or another particle with its ground state occupied. With this detection of „„ we know (or can find out)grqq, which of the particles emitted the RF photon, and it seems natural to assume that when modeling the characteristics of this selective RF instead of the amplitude $b_{12\mu}(\overline{\omega}_\mu, t)$ two of such amplitudes should be used separately: $b_{12\sigma}(\overline{\omega}_\sigma, t)$ and $b_{12\nu}(\overline{\omega}_{\nu,t})$, which reflect the evolution of each of the above states $|12\sigma\rangle$, $|12\nu\rangle$ and at $t = \infty$ have the following form:

$$b_{12\sigma}(\overline{\omega}_\sigma, t{=}\infty) = -i\hbar^{-1}\Omega\left[V_{1\sigma}^{40}(\overline{\omega}_\sigma + 2\Delta + i\gamma_3)\right]/F_\sigma(\overline{\omega}_\sigma),$$

$$b_{12\nu}(\overline{\omega}_\nu, t = \infty) = -i\hbar^{-1}\Omega V_{2\nu}^{30} w / F_\nu(\overline{\omega}_\nu), ,$$

where $\overline{\omega}_\sigma \equiv \omega_\sigma - \omega_{41}$, $\overline{\omega}_\nu = \omega_\nu - \omega_{41}$; the function $F_\sigma(\overline{\omega}_\sigma)$ has the form of the function $F_\mu(\overline{\omega}_\mu)$, in which $\tilde{\gamma} = 0$ and the frequency $\overline{\omega}_\mu$ are replaced by $\overline{\omega}_\sigma$, and the function $F_\nu(\overline{\omega}_\nu)$ has the form of the function $F_\sigma(\overline{\omega}_\sigma)$ when replacing $\overline{\omega}_\sigma$ with $\overline{\omega}_\nu$.

The given expressions $b_{12\sigma}(\overline{\omega}_\sigma, t = \infty)$ and $b_{12\nu}(\overline{\omega}_\nu, t = \infty)$ were obtained in Ref. [5] as solutions of the corresponding system of equations for the amplitudes of the basic states $|12\lambda\rangle$, $|420\rangle$, $|130\rangle$, $|12\sigma\rangle$ and $|12\nu\rangle$. The squares of the modules of the above solutions determine „non-interfering alternatives“ probabilities (see pages 25, 26 in Ref. [10]) of detecting an RF photon emitted by a particle A or a particle B with information about a certain value of the probability of occupation of the ground state of each of the particles as part of the probability of occupation of their collective ground state in case of detection of a photon of the collective RF (see the remark on this in the Introduction of Ref. [11]). It should be noted that selective RF accompanies, for example, bimolecular secondary photoreaction, since we can find out which of the molecules of the reaction center emitted a photon of RF: a reagent molecule or a molecule of the reaction product that differs from it in chemical properties. In the case of irradiation of a reagent molecule with a short pulse leading to spontaneous fluorescence of a pair of molecules of the reaction center, the dynamics of the states of the complete composite system was modeled in Ref. [12] based on the analytically obtained solutions $b_{12\sigma}(t)$ and $b_{12\nu}(t)$ of the corresponding system of equations at initial the condition $b_{420}(t = 0) = 1$.

Contours of the spectra

The RF spectrum will be called the dimensionless frequency functions of the RF photon formed by the square of the modulus of the corresponding amplitude of the steady state of the complete composite system. The RF spectrum of one isolated particle is denoted by $S_A^{RF}(\overline{\omega}_\mu)$, the spectrum of the collective RF of a pair of particles is denoted by $S_\mu(\overline{\omega}_\mu)$, the spectrum of the component of the selective RF particle A is denoted by $S_\sigma(\overline{\omega}_\sigma)$, the spectrum of the component of the selective RF particle B is denoted $S_\nu(\overline{\omega}_\nu)$. The frequency is measured in units of γ_4.

Using the above expression $b_A^{RF}(\overline{\omega}_\mu,\ t = \infty)$, when replacing the function $\Gamma_\mu(\overline{\omega}_\mu)$ by its real part (as in Ref. [8] pages 230-231) we obtain

$$S_A^{RF}(\overline{\omega}_\mu) = |b_A^{RF}(\overline{\omega}_\mu),\ t = \infty|^2$$

$$= \hbar^{-4}|V_{1\mu}^{40}|^2|V_{40}^{1\lambda}|^2/[(\overline{\omega}_\mu^2 + \gamma_4^2)[(\overline{\omega}_\mu - \overline{\omega}_L)^2 + \Gamma_\mu^2(\overline{\omega}_\mu)]$$

$$= \hbar^{-2}\Omega^2|V_{1\mu}^{40}|^2/\{[\overline{\omega}_\mu(\overline{\omega}_\mu - \overline{\omega}_L) - \Omega^2]^2 + \gamma_4^2(\overline{\omega}_\mu - \overline{\omega}_L)^2\}.$$

This function has the form of the function (20.12) in Ref. [8] and is consistent with the expression for the contour of the RF spectrum of an isolated two-level particle irradiated with monochromatic light, which was obtained in Ref. [13] see ($3''$). If we take out the multiplier $[(\omega_\sigma - \omega_0)^2 + (\gamma/2)^2]$ in the denominator of the fraction ($3''$), then after reducing this fraction, the resulting expression as a function of ϖ_σ will match the above expression $|b_A^{RF}(\varpi_\mu, t = \infty)|^2$.

To compare the contour $S_A^{RF}(\varpi_\mu)$ with the emission spectrum contour $S_A^{SF}(\varpi_\eta)$ of a spontaneously fluorescing (SF) photon η from a particle A, we express the spectrum, following Ref. [8] (see (18.13), §18, Chap. 5), as a Lorentzian profile with its maximum at $\hbar^{-2}|V_{1\eta}^{40}|^2$ (in units of γ_4^2) for $\varpi_\eta = 0$ and a half-width $2\gamma_4$:

$$|b_{1\eta}(\omega_\eta, t = \infty)|^2 \equiv S_A^{SF}(\varpi_\eta) = \hbar^{-2}|V_{1\eta}^{40}|^2/(\varpi_\eta^2 + \gamma_4^2), ,$$

where $\varpi_\eta = \omega_\eta - \omega_{41}$ and in the matrix element $V_{1\eta}^{40}$ it is assumed that $\omega_\eta = \omega_{41}$.

The condition for maintaining the normalization of amplitudes $b_{1\eta}(\omega_\eta, t = \infty)$ is the equality $\sum_\eta |b_{1\eta}(\omega_\eta, t = \infty)|^2 = 1$, in which summation is performed taking into account all equally probable directions of photon propagation η (with two states of its polarization) and all possible values of its frequency ω_η. The result of this summation looks like replacing the numerator $\hbar^{-2}|V_{2\eta}^{40}|^2$ in the above expression $b_{1\eta}(\omega_\eta, t) = \infty$ with the constant γ_4/π (see (20.9a) on page 230 in Ref. [8]) followed by integration by $d\omega_\eta$ of the resulting expression. Finally we obtain

$$\int_{-\infty}^{\infty} \gamma_4 d\omega_\eta/[\pi(\omega_\eta^2 + \gamma_4^2)] = 1..$$

Using the above expressions $b_\chi(\varpi_\chi, t = \infty)$, χ–μ, $\sigma\nu$ we obtain

$$S_\mu(\varpi_\mu) = \hbar^{-2}\Omega^2\Big\{|V_{2\mu}^{40}|^2\left[(\varpi_\mu + 2\Delta)^2 + \gamma_3^2\right])$$
$$+ |V_{2\mu}^{30}|^2(w^2 + \tilde{\gamma}^2) + 2|V_\mu^{40}||V_{2\mu}^{30}|$$
$$\times [w(\varpi_\mu + 2\Delta) - \gamma_3\tilde{\gamma}]\Big\}\Big/|F_\mu(\varpi_\mu)|^2,$$

$$S_\sigma(\varpi_\sigma) = \hbar^{-2}\Omega^2|V_{1\sigma}^{40}|^2\left[(\varpi_\sigma + 2\Delta)^2 + \gamma_3^2\right])/|F_\sigma(\varpi_\sigma)|^2$$
$$S_\nu(\varpi_\nu)\hbar^{-2}\omega^2|V_{2\nu}^{30}|^2 w^2/|F_\nu(\varpi_\nu)|^2.$$

The functions $S_\chi(\varpi_\chi)$ reflect rather complex (for analytical consideration) dependences of the position and shape of the contours of the spectra on the values of the parameters of the particles, the irradiation photon, and the RF photon in question. Therefore, the determination of such dependencies is based on the analysis of a set of graphic images (drawings) of functions $S_\chi(\varpi_\chi)$, which are obtained using yotx.ru and MATLAB 6.5 systems. As an initial general idea of the characteristic form of these dependencies, images of $S_\chi(\varpi_\chi)$ functions for individual, particular values of particle parameters and the irradiation field are considered, when choosing which conditions for photon registration by collective or selective RF seem to be the simplest. At the same time, we would like to note that it can be concluded judging by the appearance of the above expressions for the spectra that the shape of their contours is determined mainly by the type of dependence on the ϖ_χ of functions $|F_\chi(\varpi_\chi)|^2$, since the values of these functions vary in a fairly wide frequency range around $\varpi_\chi = 0$ much more than the values of the functions $|V_{1\chi}^{40}|^2$, $|V_{2\chi}^{30}|^2$ and $|V_{1\mu}^{40}||V_{2\mu}^{30}|$. Therefore, when obtaining images of contours $S_\chi(\varpi_\chi)$, it was assumed that the functions $|V_{1\chi}^{20}|^2$, $|V_{2\chi}^{30}|^2$ and $|V_{1\mu}^{40}||V_{2\mu}^{30}|$ are constant, taken at $\omega_\chi = \omega_{41}$, and are considered as parameters characterizing the structure of particles.

First of all, the drawings obtained by graphical representations of functions $S_A^{RF}(\varpi_\mu)$ are considered. In these functions, the frequency values, as noted above, are written in units of γ_4 and, accordingly, the values of the parameters $\hbar^{-2}|V_{1\mu}^{40}(\omega_\mu = \omega_{41})|^2$ and Ω^2 are written in units of γ_4^2; the value of the function $S_A^{RF}(\varpi_\mu)$ represents a positive real number for some value of its argument, so that positive real numbers are deposited along the ordinate axis (in the Cartesian coordinate system used for drawings), and along the abscissa axis — values ϖ_μ in units γ_4 (as shown in the figures in the next section of this paper).

For the case of resonant irradiation ($\varpi_L = 0$), the following conclusions are made, for example: the contour $S_A^{RF}(\varpi_\mu)$ has a maximum 10^2 at $\varpi_\mu = 0$ with half-width 2×10^{-2} with an irradiation intensity characterized by the value $\Omega = 0.1$; the contour $S_A^{RF}(\varpi_\mu)$ has two components at $\varpi_\mu = -0.7$ and $\varpi_\mu = 0.7$ with maxima equal to 1.3 with $\Omega = 1$; the contour $S_A^{RF}(\varpi_\mu)$ has two peaks with $\Omega = 10$: at $\varpi = 10$ and $\varpi = -10$ with maxima equal to 1 and half-width equal to 1.

As usual, we will use the name „weak pumping“ to irradiate a particle A with a pulse of light of such intensity that $\Omega < 1$, and „strong pumping“ — for irradiation of such intensity that $\Omega > 1$.

The following conclusion is drawn based on the type of the noted features and the totality of the obtained figures at other values of Ω: in case of weak resonant pumping of an isolated particle A, the spectral line of its RF is significantly narrower than the spectral line of its SF. In the limit $\Omega \to 0$, the contour $S_A^{RF}(\varpi_\mu)$ has the form of a Lorentzian representation approaching a δ Dirac delta function: $\delta(\varpi) = \lim_{\delta \to 0} \varepsilon/[\pi(\varepsilon^2 + \varpi^2)]$. At $\Omega \to 1$, the contour $S_A^{RF}(\varpi_\mu)$ approaches the contour $S_A^{SF}(\varpi_\mu)$, and at $\Omega \gg 1$, the contour $S_A^{RF}(\varpi_\mu)$ splits into two components with maxima equal to 1, at $\varpi = \pm\Omega$ with a half-width of 1.

Images of functions $S_A^{RF}(\varpi_\mu)$ in case of quasi-resonant irradiation ($\varpi \neq 0$) are also considered. For example, when irradiated with a frequency of $\varpi_L = 10$ at $\Omega = 0.1$, the contour $S_A^{RF}(\varpi_\mu)$ has a maximum of 10^4, which is located at $\varpi_\sigma = 10$ with a half-width of $\approx 2 \times 10^{-4}$. Based on the totality of the obtained figures, it is concluded that with weak pumping by quasi-resonant irradiation, the frequency of the maximum of the $S_A^{RF}(\varpi_\mu)$ function coincides with the frequency of irradiation, the value of this maximum is greater, and its half-width is smaller compared with

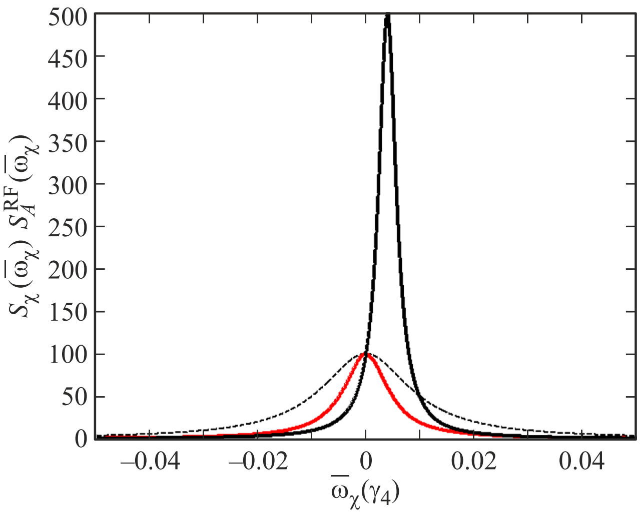

Figure 2. An example of the shape of the contours of the collective and selective RF spectra of identical, weakly interacting particles when the particle is weakly pumped by resonance irradiation (details in the text).

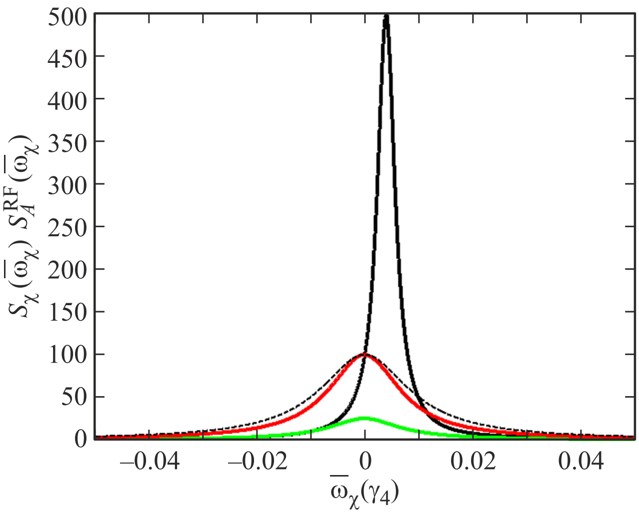

Figure 3. An example of the type of contours of RF spectra of particles with different decay of excited states of particles in case of a weak pumping of the particle by resonance irradiation.

the corresponding values of the $S_A^{RF}(\overline{\omega}_\mu)$ function under resonant irradiation. When strongly pumped, the spectrum $S_A^{RF}(\overline{\omega}_\mu)$ has two components. The maximum of one of them is located at $\overline{\omega}_\mu > \Omega$, and the other is located at such a negative frequency value $\overline{\omega}_\mu$ that $|\overline{\omega}_\mu| > \Omega$; the maximum of the first of them is slightly greater than the maximum of the second.

When using the WolframAlpha integral computation system, the fulfillment of the normalization condition was verified. Taking into account (see above) the replacement of $\hbar^{-2}|V_{1\mu}^{40}(\omega_\mu = \omega_{41})|^2$ with γ_4/π when summing over all photon propagation directions μ, this reduced to checking the equality

$$\pi^{-1}\int\limits_{-\infty}^{+\infty} S_A^{RF}(\overline{\omega}_\mu)d\overline{\omega}_\mu = 1.$$

Illustrations and discussions of the shape of the contours of the spectra

Some of the examples of images of contours $S_A^{RF}(\overline{\omega}_\mu)$ are shown with thin black shaded curves in the figures below, among the examples of images of function $S_\chi(\overline{\omega}_\chi)$. The positive real numbers are plotted along the ordinate axis in these figures (as noted above), and frequency $\overline{\omega}$ in units of γ_4 is plotted along the abscissa axis, implying that $\overline{\omega} \equiv \overline{\omega}_\mu$ when considering the function $S_\mu(\overline{\omega}_\mu)$, $\overline{\omega} = \overline{\omega}_\sigma$ when considering the function $S_\sigma(\overline{\omega})$ and $\overline{\omega} = \overline{\omega}_\nu$ when considering the function $S_\nu(\overline{\omega}_\nu)$. The function $S_\sigma(\overline{\omega}_\sigma)$ is represented by a red curve, the function $S_\nu(\overline{\omega}_\nu)$ is shown by a green curve, the function $S_\mu(\overline{\omega}_\mu)$ is shown by a black curve. Different functions $S_\chi(\overline{\omega}_\chi)$ are shown by either thin or thickened curves for visual comparison.

Images of functions $S_\chi(\overline{\omega}_\chi)$ of identical particles ($\gamma_3 = \gamma_4 = \tilde{\gamma},\ \Delta = 0$), whose interaction is characterized by different values of w, in case of resonant irradiation ($\overline{\omega}_L = 0$) of such intensity that $\Omega = 0.1$. It is assumed that the orientation of the non-parallel dipoles of the particle transition and the choice of photon parameters $\chi{\div}\mu,\ \sigma,\ \nu$ are such that, taking into account the inequality $0 \le \Delta \ll \omega_{41},\ \omega_{32}$, when considering the function $S_\mu(\overline{\omega}_\mu)$, the condition $|V_{1\mu}^{40}| = |V_{2\mu}^{30}| = \hbar\gamma_4$ can be considered valid, the condition $|V_{1\sigma}^{40}| = \hbar\gamma_4$ and $|V_{2\nu}^{30}| = 0$ can be considered valid when considering the function $S_\sigma(\overline{\omega}_\sigma)$, and the condition $|V_{2\sigma}^{30}| = \hbar\gamma_4$ and $|V_{1\sigma}^{40}| = 0$ can be considered valid when considering the function $S_\sigma(\overline{\omega}_\sigma)$. Images of such functions $S_\chi(\overline{\omega}_\chi)$ with weakly interacting particles ($w = 1$) are shown in Fig. 2.

It should be noted that in the considered case, the contour $S_\sigma(\overline{\omega}_\sigma)$ almost exactly coincides with the contour $S_\nu(\overline{\omega}_\nu)$, therefore, these contours are shown by a single red curve in Fig. 2. The maximum of the contour of the collective RF, equal to 5×10^2, is shifted by 5×10^{-3} from $\overline{\omega}_\mu = 0$ and has a half-width of 5×10^{-3}, and the maxima of the contours of the components of the selective RF are at $\overline{\omega}_\sigma = \overline{\omega}_\nu = 0$ and coincide with a maximum of $S_A^{RF}(\overline{\omega}_\mu)$, but they have a half-width twice as large. At the same time, the following equation is obtained

$$\pi^{-1}\int\limits_{-\infty}^{\infty} S_\mu(\overline{\omega}_\mu)d\overline{\omega}_\mu \equiv \overline{S}_\mu = 1,$$

$$\pi^{-1}\int\limits_{-\infty}^{\infty} S_\sigma(\overline{\omega}_\sigma)d\overline{\omega}_\sigma \equiv \overline{S}_\sigma = 0.50125,$$

$$\pi^{-1}\int\limits_{-\infty}^{\infty} S_\nu(\overline{\omega}_\nu)d\overline{\omega}_\nu \equiv \overline{S}_\nu = 0.49875.$$

It should be noted that the value $\overline{S}_\mu$ represents the population of the collective ground state of a pair of particles in a collective RF, the value $\overline{S}_\sigma$ represents the population of the ground state of a particle A, the value $\overline{S}_\nu$ represents the population of the ground state of the particle B with selective RF (as in Ref. [5]), so that the above equalities indicate that the condition for maintaining normalization of the amplitudes of the states of the complete composite system for the steady-state collective ($\overline{S}_\mu = 1$) and selective RF ($\overline{S}_\sigma + \overline{S}_\nu = 1$). By comparing the values of $\overline{S}_\sigma$ and the value of $\overline{S}_\nu$, it is possible to judge the prevalence of photon emission by a selective RF particle. The equality $\overline{S}_\mu = 1$ will not be further used to shorten the record when the equality $\overline{S}_\sigma + \overline{S}_\nu = 1$ is verified.

In the case of RF particles with $w = 5$ the maximum $S_\sigma(\overline{\omega}_\sigma)$ coincides with the maximum of $S_A^{RF}(\overline{\omega}_\mu)$, equal to 1×10^2, while the maximum of $S_\nu(\overline{\omega}_\nu)$ is equal to 2.5×10^3, and the obtained values of $\overline{S}_\sigma = 0.03865$ and $\overline{S}_\nu = 0.96135$ reflect the prevalence of radiation photon selective RF particle B; it is expressed even more strongly in the case of RF particles with $w = 10$, at which the values $\overline{S}_\sigma = 0.00995$ and $\overline{S}_\nu = 0.99005$ are obtained.

As a result of consideration of the noted and the totality of the corresponding patterns, it was concluded that the contours $S_\chi(\overline{\omega}_\chi)$ of identical particles have sharply increased maximum values and reduced half-width values compared with the corresponding contour values $S_A^{RF}(\overline{\omega}_\mu)$. The photon emission from selective RF can be attributed with approximately equal probability to both the particle A and the particle B in case of a weak particle interaction, but the probability of radiation from the particle B significantly prevails in case of a strong particle interaction.

The opposite pattern takes place in case of a quasi-resonant irradiation of identical particles. For instance, a photon of selective RF is almost completely emitted by the particle A in case of irradiation of particles with $w = 1$ with light with a frequency of $\overline{\omega}_L = 10$: the contour $\overline{S}_\sigma$ coincides with the corresponding contour $S_A^{RF}(\overline{\omega}_\mu)$ (see above). However, as shown by the graphical representations of contours $S_\chi(\overline{\omega}_\chi)$, an increase of the force of particle interaction leads to a decrease of the height of all contours $S_\chi(\overline{\omega}_\chi)$ and an increase of their half-width compared to the corresponding values of the contour $S_A^{RF}(\overline{\omega}_\mu)$; thus, for example, the values of $\overline{S}_\sigma = 0.80156$ and $\overline{S}_\nu = 0.19844$ were obtained for particles with $w = 5$, and $\overline{S}_\sigma = 0.50249$ and $\overline{S}_\nu = 0.49751$ were obtained for particles with $w = 10$, i.e. the emission of a photon by selective RF is distributed approximately equally between the two strongly interacting particles under quasi-resonant irradiation, whereas in case of a resonant irradiation of such particles (see above), the emission is strongly dominated by the particle B.

Images of contours of $S_\chi(\overline{\omega}_\chi)$ particles with different decay rates of the excited state are considered, for example, for particles with parameters $\Delta = 0$, $w = 1$, $\gamma_3 = 4$, $\gamma_4 = 1$, with weak pumping of particle A by resonance irradiation. Corresponding ratios have been adopted: $|V_{1\mu}^{40}|^2 = |V_{1\sigma}^{40}|^2$, $|V_{2\nu}^{30}|^2 = 4$, $|V_{1\sigma}^{40}||V_{\nu}^{30}| = 2$, written in units of $\hbar^2\gamma_4^2$. Expressions $\overline{S}_\sigma = 0.80008$ and $\overline{S}_\nu = 0.19992$ were obtained. The contour images of the RF spectra of such particles are shown in Fig. 3.

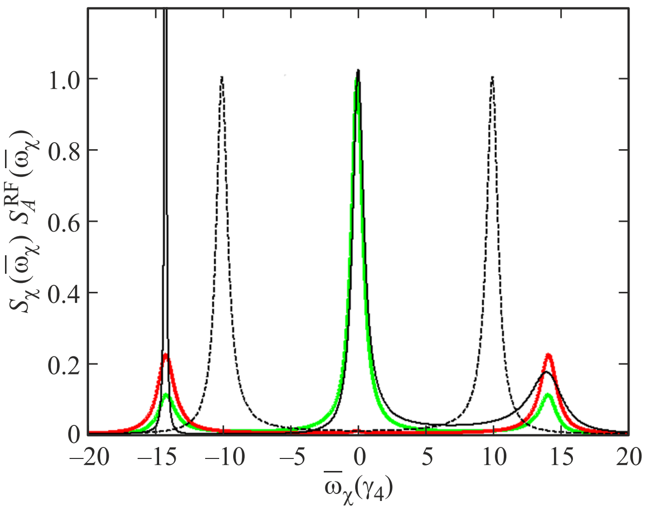

Figure 4. An example of the type of contours of RF spectra of strongly interacting identical particles in case of a strong pumping of the particle with the resonance irradiation.

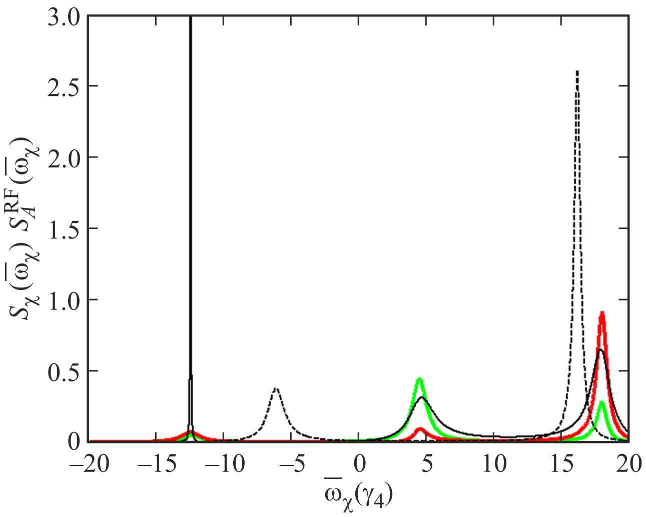

Figure 5. An example of the type of contours of RF spectra of strongly interacting identical particles when the particle is strongly pumped by quasi-resonant irradiation.

Comparing Fig. 3 with Fig. 2, we conclude that an increase of the value of γ_3 from $\gamma_3 = 1$ to $\gamma_3 = 4$ leads to a blurring of the contours of $S_\chi(\overline{\omega}_\chi)$ with a decrease observed at $\gamma_3 = 1$ of the prevalence of photon emission by a selective RF of particle A.

The study considers the images of contours $S_\chi(\overline{\omega}_\chi)$ in case of a resonant irradiation with $\Omega = 0.1$ of particles with significantly different frequencies of their proper transitions, for example, at $\Delta = 10$, $w = 1$, $\gamma_3 = 1$. Considering that $\Delta \ll \omega_{32}$, ω_{41}, the values of the matrix elements of the op-

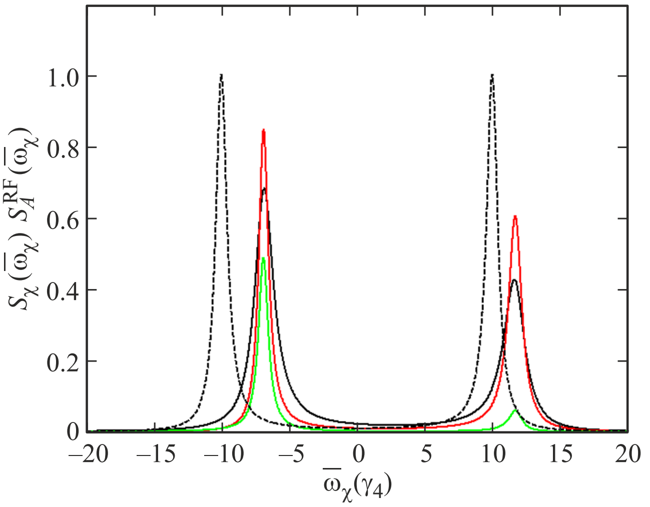

Figure 6. An example of the type of contours of RF spectra of particles with different values of their own transition frequencies when the particle is strongly pumped by resonance irradiation.

erator V for transitions between states of particles with RF photon emission are chosen the same as when considering the images of $S_\chi(\overline{\omega}\chi)$ shown in Fig. 2. In this case, the profile $S_\sigma(\overline{\omega}_\sigma)$ almost exactly replicates the profile $S_A^{RF}(\overline{\omega}_\mu)$, with a maximum 10^2 at $\overline{\omega}_{\sigma=0}$ and a half-width of 2×10^{-2}. Meanwhile, the maximum of the profile $S_\nu(\overline{\omega}_\nu)$ at $\overline{\omega}_\mu=0$ is 2×10^{-1}, with a half-width of 2×10^{-2}. The maximum of the contour $S_\mu(\overline{\omega}_\mu)$ at $\overline{\omega}_\sigma=0$ is 1.1×10^2 with a half-width of 2×10^{-2}. $\overline{S}_\sigma=0.99751$ and $\overline{S}_\nu=0.00249$ were obtained, and $\overline{S}_\sigma=0.94131$ and $\overline{S}_\nu=0.05896$ were obtained with a stronger particle interaction characterized by the value of $w=5$, so that the prevalence of the considered photon emission by the particle A decreases with the increase of the particle interaction strength.

Similar results were noted when considering the contours of RF spectra with weak pumping of a particle A by quasi-resonant irradiation.

Images of the functions $S_\chi(\overline{\omega}_\chi)$ of identical particles, the interaction of which is characterized by the value $w=1$, are considered under resonant irradiation with such intensity that $\Omega=10$. The values of the matrix elements of the operator V for transitions between states of particles with RF photon emission are chosen the same as when considering the images $S_\chi(\overline{\omega}_\chi)$ shown in Fig. 2. The spectra of $S_\mu(\overline{\omega}_\mu)$ and $S_\sigma(\overline{\omega}_\sigma)$ practically coincide with the spectrum of $S_A^{RF}(\overline{\omega}_\mu)$, which has two maxima at $\overline{\omega}_\sigma=\pm10$, equal to 1, with a half-width of 1. The spectrum $S_\sigma(\overline{\omega}_\sigma)$ has three maxima: one at $\omega=0$ and two at $\overline{\omega}=\pm10$. Each of these maxima has a height of ≈0.01. $\overline{S}_\sigma=0.98076$, $\overline{S}_\nu=0.01924$ were obtained. Each of the spectra $S_\mu(\overline{\omega}_\mu)$ and $S_{\nu(\overline{\omega}_\nu)}$ has three maxima in the case of particles with $w=10$: one at $\overline{\omega}=0$ and two at $\overline{\omega}\pm14$. The spectrum of $S_\sigma(\overline{\omega}_\mu)$ has two maxima at $\overline{\omega}_\sigma=\pm14$. $\overline{S}_\sigma=0.33775$, $\overline{S}_\nu=0.66225$ were obtained. The images of these spectra are shown in Fig. 4. For convenience of considering the type of functions $S_\chi(\overline{\omega}_\chi)$, their values along the ordinate axis are limited to 1.2, while the maximum $S_\mu(\overline{\omega}_\mu)$ for $\overline{\omega}_\mu=-14$ is 6.

As shown by the graphical images, when quasi-resonant particles are irradiated with A light with a frequency of $\overline{\omega}_L=10$ at $\Omega=10$, the photon of selective RF particles, the interaction of which is characterized by the value of $w=1$, is almost completely (as in the case of weak it is emitted by the particle A: the contour $\overline{S}_\sigma$ coincides with the corresponding contour $S_A^{RF}(\overline{\omega}_\mu)$, which has two maxima, one of which is equal to ≈0.5 at $\overline{\omega}_\mu\approx-6$ and the other is equal to ≈2.5 for $\overline{\omega}_\mu\approx16$. $\overline{S}_\sigma=0.98209$, $\overline{S}_\nu=0.01791$ were obtained, and for RF particles with $w=10$, $\overline{S}_\sigma=0.54104$ and $\overline{S}_\nu=0.45896$ were obtained, which indicates (just as in the above case of weak pumping, on the attenuation of the prevalence of the considered photon emission by the particle A. The appearance of contours $S_\chi(\overline{\omega}_\chi)$ for this case is shown in Fig. 5. For convenience of considering the type of functions $S_\chi(\overline{\omega}_\chi)$, their values along the ordinate axis are limited to 3, while the maximum $S_\mu(\overline{\omega}_\mu)$ at $\overline{\omega}_\mu=-12.5$ is 5.

Images of functions $S_\chi(\overline{\omega}_\chi)$ for particles with different decay rates of the excited state under strong pumping by resonance irradiation are considered. A conclusion has been made: just as in case of weak pumping, an increase of the value of γ_3 leads to blurring of the contours with an increase of the emission of a photon by the RF particle A.

Images of $S_\chi(\overline{\omega}_\chi)$ under resonant irradiation of particles with different frequencies of their own transitions are considered. For example, $\overline{S}_\sigma=0.99359$, $\overline{S}_\nu=0.00641$ was obtained for RF particles with parameters $\Delta=10$, $w=1$, $\gamma_3=1$ under such irradiation that $\Omega=10$, and $\overline{S}_\sigma=0.73098$, $\overline{S}_\nu=0.26902$ were obtained for RF particles with parameters $\Delta=10$, $w=10$, $\gamma_3=1$, which indicates the prevalence of RF photon emission by the particle A, as in the above case of weak pumping of particles with the same parameters. The images of these spectra are shown in Fig. 6.

It is difficult (as noted above) to write down each of the noted patterns of the relationship of spectrum characteristics with particle parameters, irradiation conditions, and recording of the RF photon in a simple analytical form. For example, the increase of the probability of photon emission by a selective RF particle B with an increase of the force of particle interaction noted by the results of consideration of graphical images of functions $S_\chi(\overline{\omega}_\chi)$ is determined not only by the fact that the resulting expression $\overline{S}_\nu$ is proportional to w^2, but also because the same expression is inversely proportional to the sum of the terms, one of which is also proportional to w^2. In such a situation, it is difficult to form a clear idea of the physical meaning of the pattern in question. Similar difficulties occur when considering other patterns of the mentioned relationship, noted as a result of consideration of graphical representations of functions $S_\chi(\overline{\omega}_\chi)$. However, establishing the physical meaning of such patterns would make a significant contribution to the theory of collective and selective interaction of interacting particles, therefore, an attempt to advance in this direction is proposed in a separate paper.

Concluding remarks

Examples of graphical images of contours of RF spectra of two interacting nanoparticles are given. The analysis of the type of these images allowed obtaining a general idea of the type of characteristic spectra under simple conditions of photon registration of collective or selective RF. It is useful to keep this idea in mind when assigning the observed contours to a particular component of the RF spectrum. The expressions obtained for the contours of the RF spectra reflect information about the dependence of the position of the contour maxima, their number, magnitude, and half-width on the values of the particle parameters, their irradiation, and detection of the RF photon. This information can be used to obtain estimates of the parameters of the structure and properties of nanoparticles based on experimental data from their RF spectra, as well as to select the values of particle parameters when considering the possibility of using them as a material for various optoelectronic devices.

Conflict of interest

The author declares that he has no conflict of interest.

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