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The effects of local environment on a probe signal absorption and resonance fluorescence for quantum emitters in transparent media

© E.A. Smirnova[1,2,3], N.A. Lozing[1,2,3], M.G. Gladush[1,3], A.V. Naumov[1,3,4]

[1] Institute of Spectroscopy, Russian Academy of Sciences, 108840 Moscow, Russia
[2] National Research Institute„Higher School of Economics ", 101100 Moscow, Russia
[3] Moscow Pedagogical State University, 119435 Moscow, Russia
[4] Troitsk separate subdivision of LPI RAS, 142190 Moscow, Russia

e-mail: ksmirno@yandex.ru, Lozing@phystech.edu, mglad@isan.troitsk.ru

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The current paper demonstrates theoretical analysis of two types of spectral curves for several configurations of system of two-level light emitters, considering the influence of local field and close environment inside a transparent medium. Probe field absorption spectra and resonant fluorescence spectra are calculated under excitation of a strong monochromatic cw laser. The sensitivity of absorption and emission optical spectroscopy method is compared for revealing the effects of the medium on individual emitters and their ensembles. Spectral curves were calculated for model emitters considering local field influence of a transparent dielectric medium and local electron-phonon interactions, which determined the response of the emitters to an external laser field and effective relaxation mechanisms. The calculation formalism is based on a semiclassical approach, while the relaxation processes associated with the phonon contribution are introduced phenomenologically with references to other studies.

Keywords: absorption spectrum, resonance fluorescence, probe field method, quantum dot, electron-phonon interaction, local field.

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1. Introduction

In modern studies and applications light can serve as a source of information about the object emitting it and, conversely, be a tool for desired or selective action on the object. In both cases it is necessary to consider light-dependent processes developing in the macro-, micro-, and nanoscales. The light absorption by individual particles and their ensembles, followed by the emission of secondary radiation (photoluminescence), combines these issues within the framework of one problem. In this case, emitters-impurities are of particular interest. The introduction of photoactive particles into a transparent material ensures a stable spatial distribution of quantum emitters and, as a consequence, a certain nature of the photoluminescence produced by them. It is known that the characteristics of the secondary radiation are determined by a combination of the properties of the carrier and the particles themselves. Some types of emitters-impurities can exhibit the phenomenon of resonant fluorescence, that is a situation where photoluminescence is provided by only two states of the quantum emitter. Atoms in the field of a laser wave close in frequency to an individual atomic transition can certainly act as two-level emitters. The resonant fluorescence of independent single particles should demonstrate rich distinctive properties, which, first of all, include photon antibunching effects and a three-peak emission spectrum [1,2]. Specifically these features were repeatedly observed for some organic molecules [3], semiconductor quantum dots [4,5] and color centers in diamonds [6]. Thus, resonant fluorescence is inherent in many types of quantum emitters under a number of specific conditions. It is important to note that all of these quantum particles alternative to natural atoms can interact with light in the mode of a two-level emitter exactly under conditions when the emitter is inside the carrier. Modern technologies make it possible to fix particles in solid matrices or on surfaces of solids. However, the location of individual particles within the medium makes them unequal in one or more parameters due to the inhomogeneities of the carrier and the presence of local conditions. Therefore, the specific location of the emitter determines what kind of light it emits. On the one hand, this makes ensembles of emitters inhomogeneous, and on the other hand, it makes it possible to use such emitters as sensors that detect various phenomena and states of the medium at different scales. The last problem can be solved by analyzing data obtained by optical spectroscopy. At the same time, optical spectroscopy makes it possible to study the properties of quantum systems, and for some material structures it may be the only available type of measurement. In this paper we present a computational-theoretical analysis of resonant fluorescence spectra and absorption spectra of a probe field for various two-level emitters with different channels of interaction between themselves and with the medium. In this study we consider the spectra for independent set of single two-level emitters, a collective ensemble of interacting two-level emitters, an independent set of single model quantum dots considering the electron-phonon interaction, and also an ensemble of such model quantum dots.

The phenomenon of resonant fluorescence has generalizations to more complex cases, i.e. it can be relevant for emitters with a system of many levels and presence of more than one resonant transition [7,9]. At the same time, the resonant fluorescence spectrum is always specific, which is used to determine the presence of the phenomenon itself. In the limit of a „strong field“, i.e. under conditions of the transitions saturation or close to it, the observer will fix the presence of additional maxima with respect to the absorption spectrum or the natural emission contour. This phenomenon is also known as the dynamic Stark effect [10] and the formation of „dressed atomic states“ [11]. Thus, the inelastic component of the resonant fluorescence of two-level emitter has a three-peak structure, known as Apanasevich–Mollow triplet [12,13]. The triplet contains a central peak at the frequency of the exciting laser (Rayleigh scattering component) and two satellites tuned in the region of lower and higher frequencies. The frequency distances between the peaks, the ratio of their heights and widths are determined by the effective values of the transition parameters of the emitter that excites light, the processes of dephasing and energy exchange with the carrier matrix. Specific resonant fluorescence spectra were first observed in an experiment with a collimated beam of sodium atoms [1]. This observation was followed by numerous experiments with atomic gases, which fully confirmed the earlier theoretical descriptions and confirmed that atoms can behave as „ideal“ two-level emitters, including the manifestation of the photon antibunching effect [2]. It took several decades to make it possible to observe resonant fluorescence with other types of emitters. At present, resonant fluorescence in the visible range has been successfully observed on single molecules [3] and semiconductor quantum dots [4,5,14]. Organic molecules can also be considered as two-level systems when interacting with each other. This is observed in experiments on the photoluminescence registration from a pair of closely spaced molecules with dipole-dipole interaction [15]. In this case, the entangled cooperative system will have four energy states and a complex fluorescence spectrum [3]. In this case, the photoluminescence excitation spectrum will reflect the fact of excitation of three collective states and form a cooperative triplet. Similarly to the analysis of the Mollow triplet, the state of emitters and their environment can be restored from the characteristics of three maxima in the spectral dependence. In this case, note that the triplet of photoluminescence of two-level particle and the photoluminescence excitation triplet of a cooperative pair of two-level particles are united by the spectral equidistance of the maxima and the presence of regularities that determine the ratios of their heights and widths.

In addition to recording the photoluminescence emission and excitation spectra or determining the statistics of photons from two-level emitters, it may also be useful to study the effective two-level medium under the influence of two tunable lasers. In this case, one of the lasers will provide pumping of emitters with a controlled power and a set resonance detuning. The second beam represents a scanning probe signal from a low power tunable laser system. The pump laser creates a „dressed“ system of light and two-level system for a specific power and detuning. Then, the absorption or amplification of the probe signal is measured depending on its frequency (wavelength). This function will provide information about the structure of dressed states [11,16] produced by the pump laser. At frequencies corresponding to the position of the satellites of the Mollow triplet, the probe signal will either be absorbed or amplified in maximum extent depending on the sign of the pump detuning from the „atomic“ transition. The probe field amplification occurs due to the energy of the pump laser [16]. The position of the absorption and amplification bands, in turn, can be controlled by varying the pump power. Combinations of these spectroscopic techniques are proposed in this article as tools for determining the two-level behavior of real emitters and studying the influence of relaxation processes, cooperative effects, and interactions with the phonon reservoir on their absorption and emission properties. The probe field absorption spectroscopy method will be implemented theoretically to calculate the absorption spectra of a weak probe signal and resonance fluorescence spectra. This method of calculating the spectral characteristics of radiation belongs to completely semiclassical approaches in the presence of a known master equation for the density matrix of the emitter.

2. Master equation for emitter and equations for spectral characteristics

To calculate the characteristics of the emitted light, it is necessary to determine the nature of the work of the material system, which is a source of light and an absorber of external radiation. As a basis, let us consider an ensemble of two-level emitters distributed in a material that weakly absorbs incident external light. The carrier material will be described as a continuous medium that fills the space between the emitters and is characterized by a complex function of the dielectric permeability. In the first approximation, when constructing the model, assume that the ensemble represents a set of identical particles uniformly distributed in the volume of the sample. For an individual particle in such an ensemble, the master equation for its density matrix ρ was obtained in [17–19]. The derivation of this constitutive equation did not require the use of phenomenological procedures and was carried out in the framework of the multiparticle quantum-kinetic formalism based on Bogolyubov chains of reduced density matrices and correlation operators. In the present analysis this master equation is used to introduce and analyze components that describe the effect of the medium on the emitter. According to [19], in general in the interaction representation for the density matrix ρ_I, one can write

$$i\,\frac{d}{dt}\rho_I = -\frac{l(\epsilon)}{\hbar}\left[\hat{\mathbf{d}}_I\left(\mathbf{E} + \int\limits_0^t dt' \sum_{a'\neq a} \overleftrightarrow{\mathbf{G}}_{aa'}\mathbf{p}_{a'}\right), \rho_I\right] - \frac{l(\epsilon)}{\hbar}\left[\hat{\mathbf{d}}_I, \int\limits_0^t dt' \left(\overleftrightarrow{\mathbf{G}}^{+}_{aa}\hat{\mathbf{d}}_I\rho_I + \overleftrightarrow{\mathbf{G}}^{-}_{aa}\rho_I\hat{\mathbf{d}}_I\right)\right]. \tag{1}$$

Here, $\hat{\mathbf{d}}_I = \mathbf{f}(t)\hat{\sigma}^+ + \mathbf{f}^*(t)\hat{\sigma}^-$ is operator of dipole moment of transition, where $\mathbf{f}(t) = \mathbf{u}\mu\exp(i\omega_0 t)$, $\hat{\sigma}^+$, and $\hat{\sigma}^-$ are atomic raising and lowering operators. The function $\mathbf{f}(t)$ consists of a unit vector in the direction of the dipole $\mathbf{u}$, matrix element for the electric dipole transition μ, and a time dependence with the transition frequency $\omega_0 = (E_2 - E_1)/\hbar$, where E_1 and E_2 are the energies of the ground and excited states of the emitter, respectively. The first part of the field $\mathbf{E} = \mathbf{E}(\mathbf{r}_a, t)$ is the solution for a plane monochromatic wave in a medium with dielectric permeability ϵ at the point $\mathbf{r}_a$ corresponding to the emitter position. The notation $\overleftrightarrow{\mathbf{G}}_{aa'} = \overleftrightarrow{\mathbf{G}}(\mathbf{r}_a - \mathbf{r}_{a'}, t - t')$ corresponds to Green tensor for field in this medium with top indexes ± corresponding to advanced and retarded component of tensor, respectively, and $\overleftrightarrow{\mathbf{G}}^{\pm}_{aa} = \overleftrightarrow{\mathbf{G}}^{\pm}_{aa}(\mathbf{r}_a - \mathbf{r}_a, t - t') = \overleftrightarrow{\mathbf{G}}^{\pm}(0, t - t')$. The non-operator values $\mathbf{p}_{a'} = \langle\hat{\mathbf{d}}\rangle_{a'}$ are the induced dipole moments of the remaining emitters in the ensemble, where $\langle\ \rangle$ designates the quantum mechanical average. The function $l(\epsilon)$ is the correction factor for the local field created by the carrier matrix, which is equal to $l(\epsilon) = 1$ for $\epsilon = 1$. This function takes one of the explicit forms depending on the model chosen for estimating the local field introduced by the carrier matrix [17]. The remaining notations are standard and refer to time, the imaginary unit, Planck constant, and the commutator of operators. In this paper equation (1) will be the base for derivation of all subsequent equations for the systems under consideration. Comments on its components will be given below as needed. In general, upon permitting the integration over time and summation over ensemble particles in the rotating wave approximation equation (1) will provide description of the emitter density matrix taking into account local fields introduced by the ensemble and the continuous medium and effective radiative relaxation rates. Thus, all known parameters of constitutive equations, such as shifts of natural frequencies, pumping and relaxation rates, will depend on the parameters of the carrier. For the purposes of this work, the possibility of such changes without discussing the specifics of exact experimental situations is of interest.

The spectra calculation method within the framework of the semiclassical description can be explained by the example of the simplest case of the Lindblad master equation or the Bloch optical equations. Calculating the emission and absorption of systems with a more complex master equation would require similar, computationally intensive steps. Let us show that equation (1) in the absence of medium and other emitters leads exactly to the basic equations for two-level atom with continuous pumping. For the case of a single emitter in the ensemble located at the point $\mathbf{r}_a$, the component with the sum in the first commutator disappears. By applying the Markov approximation to the density matrix in the integrand in the second commutator, one can easily perform integration by time. In the rotating wave approximation this component gives the radiative relaxation and the radiative shift of the transition frequency. Two contributions to the equation are defined respectively by the imaginary and real parts of the expression. Since the frequency shift is usually assumed to be small then it is sufficient to leave the imaginary part only. Then, it is necessary to pass to the Schrëdinger picture for the density matrix ρ using the energy operator of free emitter $\hat{H}_0 = \hbar\omega_0\sigma_z$, which is determined by the population inversion operator $\sigma_z = 1/2[\sigma^+, \sigma^-]$. Operator of dipole transition now will be $\hat{\mathbf{d}} = \mathbf{u}\mu(\sigma^+ + \sigma^-)$. Besides, as already noted, it makes sense to temporarily simplify the problem by considering medium with $\epsilon = 1$. Here and in next models, field of incident radiation at the frequency ω always will be described as $\mathbf{E}_d = \mathbf{e}E_d(t) = \mathbf{e}\mathscr{E}\,\mathrm{Re}\{\exp(i\omega t)\}$ with amplitude $\mathscr{E}$ and single polarization vector $\mathbf{e}$. After completing these steps, we will receive the well-known equation:

$$i\,\frac{d}{dt}\rho = \frac{1}{2}\,\omega_0[\sigma^+\sigma^- - \sigma^-\sigma^+, \rho] - \frac{1}{\hbar}\,\mathbf{u}\cdot\mathbf{e}\mu E(t)[\sigma^+ + \sigma^-, \rho] + \frac{i}{\hbar^2}\,\mu^2(\mathbf{u}\cdot\mathrm{Im}\,\overleftrightarrow{\mathbf{G}}^{\omega_0}_{aa}\cdot\mathbf{u})\big([\sigma^-, \rho\sigma^+] - [\sigma^+, \sigma^-\rho]\big), \tag{2}$$

where $\overleftrightarrow{\mathbf{G}}^{\omega_0}_{aa} = \overleftrightarrow{\mathbf{G}}(\mathbf{r}_a - \mathbf{r}_a, \omega_0) = \overleftrightarrow{\mathbf{G}}(0, \omega_0)$. Equation (2) can be rewritten in a more compact and recognizable form by making a few transformations and introducing traditional notation. The first component on the right side is defined by the free atom energy operator $\hat{H}_0$. In the second component for induced dipoles we get $(\mathbf{u}\cdot\mathbf{e}) = 1$ and can write down interaction operator $\hat{V} = -\hbar\Omega(e^{i\omega t} + e^{-i\omega t})(\sigma^+ + \sigma^-)$, where $\Omega = \mu\mathscr{E}/2\hbar$ is Rabi frequency. The last component is the radiative decay with the rate $\gamma = 2\mu^2/\hbar\mathbf{u}\cdot\mathrm{Im}\,\overleftrightarrow{\mathbf{G}}(0, \omega_0)\cdot\mathbf{u} = 4\mu^2\omega_0^3/3\hbar c^3$, where c is the light speed in vacuum. Operator part after expansion of commutators forms exactly well-known Lindblad operator $\hat{\mathscr{L}}_{\hat{O}}(\rho) = 2\hat{O}\rho\hat{O}^\dagger - \hat{O}^\dagger\hat{O}\rho - \rho\hat{O}^\dagger\hat{O}$, where in given case the arbitrary operator $\hat{O}$ is replaced by operator σ^-.

Equations (1) and (2) describe only natural radiative decay/relaxation, since (1) was obtained for stationary particles. However, any impurity emitter is characterized by the process of dephasing. To consider this circumstance, (2) to be supplemented by known operator $\hat{D}(\rho) = 2\sigma_z\rho\sigma_z - \sigma_z\sigma_z\rho - \rho\sigma_z\sigma_z$, where $\gamma_\perp$ is the rate of transverse relaxation. Determining the process rate $\gamma_\perp$, one can formally consider the influence of all dephasing processes that occur in ensembles and carriers. If we collect all relaxation processes in one operator

$$\hat{R}(\rho) = \frac{\gamma}{2}\hat{\mathscr{L}}_{\sigma^-}(\rho) + \gamma_\perp \hat{D}(\rho),$$

then the master equation is transformed to the form well-known in the literature:

$$i\,\frac{d}{dt}\,\rho = \frac{1}{\hbar}\,[\hat{H}_0 + \hat{V}, \rho] + i\hat{R}(\rho). \tag{3}$$

Such a record of the equation will be convenient for describing the method of probe field spectroscopy. Formally, it will be necessary to consider the case when the external field in equations (1), (2) contains both strong and weak monochromatic components. It is convenient to present the weak component as $\mathbf{E}_p = \mathbf{e}_p E_p(t) = \mathbf{e}_p \mathscr{E}_p \mathrm{Im}\{\exp(i\omega_p t)\}$, for which $\mathscr{E}_p \ll \mathscr{E}$. Then in final equation (3) additional commutator with operator $\hat{V}_p = i\hbar\lambda_p(e^{i\omega_p t} - e^{-i\omega_p t})(\sigma^+ + \sigma^-)$ shall arise. The density matrix defined by the new equation with two fields will be denoted as ϱ. Further, we will assume that the frequency ω_p is tuned close to the transition frequency ω_0. The coupling coefficient $\lambda_p = (\mu\mathscr{E}_p)/2\hbar$ is determined by the amplitude of the weak field component $\mathscr{E}_p$ and is always very small as compared to the Rabi frequency determined by the strong field. Since this approach is a method for calculating the spectrum and not a problem of interaction with a non-monochromatic field, it is assumed that the issues of phase and polarization are left behind the scope. In this configuration, we can find solution for the density matrix ϱ in the form $\varrho = \rho + p$, where p is a small correction to the matrix, which comply with equation (3) for the case of single strong field. Then, the equation for $\rho + p$ takes the form

$$\begin{aligned} i\,\frac{d}{dt}\,(\rho + p) &= \frac{1}{\hbar}\,[\hat{H}_0 + \hat{V}, \rho + p] \\ &\quad + \frac{1}{\hbar}\,[\hat{V}_p, \rho + p] + i\hat{R}(\rho) + i\hat{R}(p). \end{aligned} \tag{4}$$

Subtracting equation (3) from equation (4) and neglecting the component of the second order of smallness, i.e. $[\hat{V}_p, p]$, we obtain the equation for the correction matrix p:

$$i\,\frac{d}{dt}\,p = \frac{1}{\hbar}\,[\hat{H}_0 + \hat{V}, p] + i\hat{R}(p) + \frac{1}{\hbar}\,[\hat{V}_p, \rho]. \tag{5}$$

The system of linked equations (3) and (5) will be the basis for further analysis. Before obtaining from them a system of kinetic equations for the components of the matrices $\rho_{kl}(t)$ $(k, l = 1, 2)$ and $p_{mn}(t)$ $(m, n = 1, 2)$, they must be reduced to equations with time-independent parameters. This can be done using next substitutions $\rho_{21} = r_{21}e^{-i\omega t}$, $\rho_{12} = r_{12}e^{i\omega t}$ which lead (3) to system rotating with frequency of laser, and $p_{21} = \phi_{21}e^{-i\omega_p t}$, $p_{11} = \phi_{11}e^{i\nu t}$, $p_{22} = \phi_{22}e^{i\nu t}$, $p_{12} = \phi_{12}e^{i(2\omega - \omega_p)t}$, where $\nu = \omega - \omega_p$. After making these substitutions, the rapidly oscillating terms can be neglected, which will lead to the final system of equations:

$$i\,\frac{d}{dt}\,r = \frac{1}{\hbar}\,[\hat{H}'_0 + \hat{V}', r] + i\hat{R}(r), \tag{6}$$

$$i\,\frac{d}{dt}\,\phi = \nu\phi + \frac{1}{\hbar}\,[\hat{H}'_0 + \hat{V}', \phi] + i\hat{R}(\phi) - i\lambda_p[\sigma^+, r], \tag{7}$$

where r and ϕ are new matrices, and $\hat{H}'_0 = \Delta\sigma_z$ and $\hat{V}' = -(\beta\sigma^- + \beta^*\sigma^+)$ are components of Hamiltonian written in compliance with approach of the rotating wave. The part corresponding to the free particle is now proportional to $\Delta = \omega_0 - \omega$, which describes the detuning between the transition frequency and the strong field. As for the interaction operator, it now contains a multiplier, which in the simple case under consideration is equal to $\beta = \beta^* = \Omega$ and will take a more complex form under other circumstances.

It is important to note that both matrices ϱ and ρ retain their fundamental properties, i.e. $\mathrm{tr}(\varrho) = \mathrm{tr}(\rho) = 1$. Therefore, the correction matrix must satisfy the condition $\mathrm{tr}(p) = 0$. As is easy to see that the same applies to the matrices r and ϕ. Averages of operators are calculated according to $\langle\hat{O}\rangle = \mathrm{tr}(\hat{O}\rho + \hat{O}p) = \langle\hat{O}\rangle_{strong} + \langle\hat{O}\rangle_{weak}$. So, contribution of the probing signal in general response of particles to the external field action shall be evaluated using component of the induced polarization $\mathbf{P}_p = N\langle\hat{\mathbf{d}}\rangle_{weak} = \mathbf{e}_p N 2\mu \mathrm{Re}(p_{21})$.

From Maxwell equations it follows that volumetric specific power of field, which creates electrical polarization of medium, is equal to $\langle\mathbf{E}\cdot\partial\mathbf{P}/\partial t\rangle_t$, where $\langle\ \rangle_t$ denote averaging by time. Since we need to find a stationary solution, we need to set $dr/dt = d\phi/dt = 0$, while the fraction of the probe signal can be calculated as

$$W_{weak} = \left\langle \mathbf{E}_p \cdot \frac{\partial\mathbf{P}_p}{\partial t} \right\rangle_t = -N\mu\mathscr{E}_p\omega_p \,\mathrm{Re}(\phi_{21}). \tag{8}$$

On the other hand, this value can also be defined as the change in signal intensity per unit length along the propagation axis, i.e. we can write

$$N\mu\,\mathscr{E}_p\omega_p \,\mathrm{Re}(\phi_{21}) = \frac{dI_p}{dz} = -\alpha I_p,$$

where I_p is the intensity of the probe field, which can be expressed as follows $I_p = (c\mathscr{E}_p^2)/8\pi$. Finally, the absorption coefficient of the probe field at a certain frequency is equal to

$$\alpha = -\frac{4\pi}{\hbar}\,N\mu^2\,\frac{\omega_p}{c}\,\mathrm{Re}\left(\frac{\phi_{21}}{\lambda_p}\right). \tag{9}$$

Next, to find ϕ_{21}, it is necessary to solve equations (6) and (7) for the stationary case. For this, it can be convenient to pass to new real functions from combinations of components of the matrix r. For this rewrite r into basis of Bloch vector $r = 1/2(\mathbf{I} + \mathbf{B}\cdot\sigma)$, where $\mathbf{I}$ is unit matrix, $\mathbf{B}^T = \{r_{21} + r_{12}, r_{11} - r_{22}, i(r_{12} - r_{21})\} = \{u, w, v\}$ is Bloch vector, and $\sigma^T = \{\sigma_1, \sigma_2, \sigma_3\}$ is vector composed from Pauli matrices. The equations that follow from equation (6) lead to known stationary solutions. For simplicity, we limit the problem to the case when, in the absence of perturbations, the system passes to its ground state, while there are no true restrictions on the introduction of equilibrium thermal populations. Thus, set $\mathbf{B}_{eq}^T = \{0, w_{eq}, 0\}$, where $w_{eq} = 1$.

It is convenient to write solutions for components $\phi(\nu)$ in stationary case in form of vector $\boldsymbol{\varphi}^T(\nu) = \{\phi_{21}, \phi_{11}, \phi_{12}\}$. Also, remember that $\phi_{22} = -\phi_{11}$ is true due to the property described above. Thus, the solution ϕ is found from the equation:

$$\mathbf{M}(\nu)\boldsymbol{\varphi} - \mathbf{s} = 0, \tag{10}$$

where

$$\mathbf{M}(\nu) = \begin{bmatrix} \nu + \Delta - i\gamma_2 & -2\beta^* & 0 \\ -\beta & \nu - i\gamma & \beta^* \\ 0 & 2\beta & \nu - \Delta - i\gamma_2 \end{bmatrix} \tag{11}$$

and the vector $\mathbf{s}$ is determined by the inhomogeneity $i\lambda_p[\sigma^+, r]$. The decay rate γ_2 can describe both the dephasing $\gamma_2 = \gamma_\perp$ and $\gamma_2 = \gamma/2$ in the case of pure radiative decay. The vector $\mathbf{s}$ is written in terms of the Bloch vector components as follows:

$$\mathbf{s} = \frac{i}{2} \begin{bmatrix} 2w \\ u + iv \\ 0 \end{bmatrix}. \tag{12}$$

Now the absorption coefficient of the probe signal (10) is found from

$$\alpha(\nu) = -\frac{4\pi}{\hbar} N\mu^2 \frac{\omega_p}{c} \operatorname{Re}\big(\mathbf{M}^{-1}(\nu)\mathbf{s}\big)_1. \tag{13}$$

To obtain the emission (fluorescence) power spectrum, one needs to transform the inhomogeneous term in equation (7). Thus, here, when determining the radiation power, we will follow the concept developed and described in [16]. Indeed, the operation of the probe signal should be considered in relation to transitions in a controlled quantum system described by r matrix. If „manually“ exclude the role of the ground state from the last component in equation (7) and introduce a new solution into equation (8), then the latter, taken with the opposite sign, should describe the specific power transferred to the field from the emitters. Since the transmitted power contains both an elastic component, i.e. pure Rayleigh scattering of the external field, and an inelastic component, it will be interesting for us to exclude the elastic part in advance. These „artificial“ procedures look as follows:

$$i\lambda_p[\sigma^+, r] \rightarrow -i\lambda_p(r\sigma^+ - r\langle\sigma^+\rangle). \tag{14}$$

This means that only a part of the commutator in the inhomogeneous term of equation (7) is taken into consideration, but at that the raising operator is also replaced by its difference with its average value. In [20] the legitimacy of such a subtraction was proved rigorously and in detail within the framework of a fully quantum-mechanical description. Finally, putting together (7), (8), (10), (11) and (14), we receive a new vector:

$$\mathbf{s}_f = -\frac{i}{2}\lambda_p^2\left(\begin{bmatrix} 1 - w \\ u + iv \\ 0 \end{bmatrix} + (u + iv) \begin{bmatrix} u - iv \\ 1 + w \\ u + iv \end{bmatrix} \right), \tag{15}$$

which must be used to calculate the contribution of one emitter to the power transferred to the field:

$$W_f(\nu) = 2\hbar\omega_p \operatorname{Re}\big(\mathbf{M}^{-1}(\nu)\mathbf{s}_f\big)_1. \tag{16}$$

Equations (13) and (16) are the final formulas for calculating the functions that describe the absorption and emission characteristics in this study. The results of calculations of the spectral dependences for single emitter without environment are shown in Figs. 1, *a* and 2, *a*, respectively. All dependences $\alpha(\nu)$ and $W_f(\nu)$ are presented in relative units (dimensional coefficients), while all parameters are expressed in units of the radiative decay rate γ. Figure 1, *a* shows the absorption spectra for three combinations of detuning of the strong exciting field from the transition frequency of the emitter and the value of the control Rabi frequency. For similar conditions, the Mollow triplets in Fig. 2, *a* were calculated. It follows from the comparison of the Figures that both spectroscopic techniques can be effective and informative for analyzing the photoluminescence of two-level emitters. If the experimental observation of the Mollow triplet requires the availability of spectral devices and high-resolution techniques, then for probe field spectroscopy it is necessary to determine the absorption and amplification bands by tuning the frequency of the probe laser. The contributions of various additional processes affecting the emitter can be determined from spectral positions, relative magnitudes, and spectral peak widths.

3. Modifications of spectra of two-level emitters

Let us consider how some types of interactions of the emitter with its local environment in the carrier matrix can modify the contours of the spectral dependences. The influence of local fields and local vibrational modes stands out the strongest influences on the optical properties of particles. At that, the local fields are due to the dipole-dipole interaction of the emitter with other emitters and components of the medium. Also, a continuous medium imposes conditions on the field structure, which leads to the Parcel effect occurrence [17,21]. At that, these effects can provide a feedback between effective pumping, decay, and the state of the emitter.

Let us now return to equation (1) and reproduce all transformations for system of N emitters per unit volume located at distances smaller than the excitation/emission wavelength. In a dense homogeneous ensemble excited by laser light, each atom is actually under the action of effective (Lorentz) local field $\mathbf{E}_\mathrm{L}$, which consists of an external field $\mathbf{E}$ and macroscopic polarization $\mathbf{P}$. This field can be written in terms of the elements of the density matrix [20,21]

$$\mathbf{E}_\mathrm{L} = \mathbf{E} + \frac{4\pi}{3}\mathbf{P} = \mathbf{E} + \frac{4\pi}{3} N\mathbf{u}\mu\,(\rho_{12} + \rho_{21}),$$

where N is concentration of emitters. Taking into account the effective value of $\mathbf{E}$, we obtain equation (6), where the value β in the matrix $\mathbf{M}$ takes the effective value:

$$\beta = \Omega + \left(\varepsilon + \frac{i\gamma_c}{2}\right)(u - iv). \tag{17}$$

Here, $\varepsilon = 4\pi N\mu^2/3\hbar$ is the so-called Lorentz red shift and $\gamma_c = N\gamma/2$ is the collective decomposition rate [20]. It follows from this that the local field consideration leads to a change in the matrix $\mathbf{M}(\nu)$. Dynamic components appear inside it, depending on the state of the emitter. In other words, the values of the elements of the emitter density matrix now determine not only the matrices (12) and (15), but also the main matrix (11). It follows from the structure of the latter that dynamic changes β will lead to shifts in the absorption/amplification zones of the probe signal and shifts in the satellites of the resonant fluorescence spectrum. However, unlike single free emitters (Fig. 1, *a* and 2, *a*) their distance from the center will no longer be proportional to the Rabi frequency $\beta = \Omega$. This effect is shown in Figs. 1, *b* and 2, *b*. It can be seen that, as a result of the action of collective local fields, the pumping increasing (doubling Ω) leads only to insignificant shift of the spectral lines (compare with Figs. 1, *a* and 2, *a*). At the same time, for the same pumping, the Lorentz red shift increasing significantly changes the position of the shifted spectral components. It can be seen that the widths of the lines and the ratio of their heights also change. However, it can be assumed that the effect of width change will not be so noticeable as compared to the widenings introduced by dephasing and interaction with phonon modes. For the types of emitters under consideration this is an important circumstance and should be specified or considered when modeling.

To construct spectra (13) and (16) for two-level emitters with strong dephasing and considering the phonon contribution, we use the master equation obtained in the paper [4]. This paper presents theoretical and experimental studies of the resonant fluorescence of semiconductor quantum dots, which were prepared for operation in the dressed state mode of a two-level system. The master equation in this case took the following form:

$$\begin{aligned} i\frac{d}{dt}\rho = \frac{1}{\hbar}[\hat{H} + \hat{V}, \rho] + i\hat{R}(\rho) + i\gamma^{+}\hat{\mathscr{L}}_{\sigma^{+}}(\rho) \\ + i\gamma^{-}\hat{\mathscr{L}}_{\sigma^{-}}(\rho) - i\gamma_{cd}(\sigma^{+}\rho\sigma^{+} + \sigma^{-}\rho\sigma^{-}), \end{aligned} \tag{18}$$

where γ_{cd} is the cross dephasing rate that affects the off-diagonal elements of the density matrix, the component with γ^- corresponds to enhanced radiative decomposition, while the component containing γ^+ is incoherent excitation process. The parameters $\gamma^{\pm}$, γ_{cd} have a slow dependence on the detuning with the laser frequency and can be considered as constants within the limits of the used pump frequencies. Thus, this equation describes a two-level system that interacts with a light source and a phonon reservoir, and also considers the processes of cross-relaxation of the diagonal elements of the density matrix.

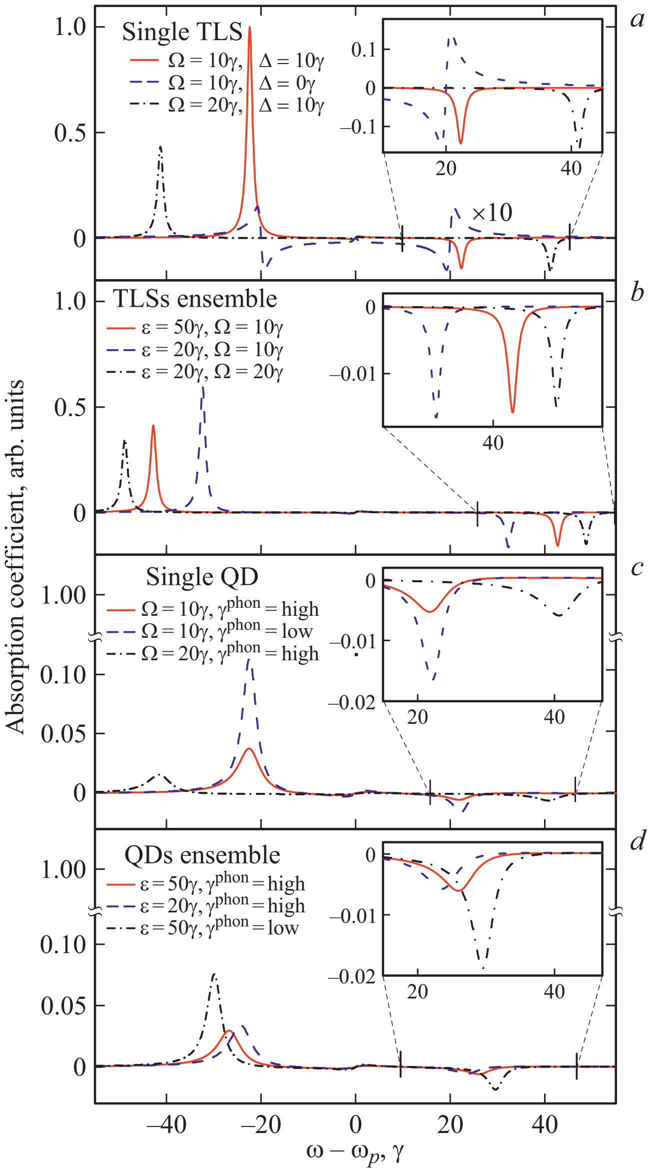

Figure 1. Spectrum of absorption of weak probe signal for various parameters: *a*) single two-level emitter; *b*) ensemble of two-level emitters (here, $\Delta = 10\gamma$, $\gamma_c = 0.9\gamma$); *c*) model QD (here, $\Delta = 10\gamma$), where „$\gamma^{phon} = high$“ denotes set of parameters $\gamma^+ = 0.9\gamma$, $\gamma^- = 0.8\gamma$, $\gamma^{cd} = 0.6\gamma$ and „$\gamma^{phon} = low$“ denotes $\gamma^+ = 0.45\gamma$, $\gamma^- = 0.4\gamma$, $\gamma^{cd} = 0.3\gamma$; *d*) ensemble of QD (here, $\Omega = 10\gamma$, $\Delta = 10\gamma$, $\gamma_c = 0.9\gamma$).

Note that equation (18) coincides with equation (3) in the first two components. New components change the matrix $\mathbf{M}$, they are represented by introducing a new matrix $\mathbf{M}_{pn}$, which is the sum of the matrix defined in (11), and an additional matrix:

$$\mathbf{M}_{pn}(\nu) = \mathbf{M}(\nu) + \\ + \begin{bmatrix} -i(\gamma^+ + \gamma^-) & 0 & -i\gamma_{cd} \\ 0 & -2i(\gamma^+ + \gamma^-) & 0 \\ -i\gamma_{cd} & 0 & -i(\gamma^+ + \gamma^-) \end{bmatrix}. \tag{19}$$

Let us first consider the case when the pumping in the matrix $\mathbf{M}$ is determined by the external field only, i.e. $\beta = \beta^* = \Omega$. Next, we will use the matrix $\mathbf{M}_{pn}$ and calculate using formulas (13) and (16). The resonance fluorescence spectra become asymmetric with respect to the heights of the satellites (Fig. 2, *c*), which exactly reproduces the result of the paper [4]. At that in this paper, we present a calculation of the absorption spectra of the probe field, from which one can see a clear weakening of the absorption/amplification effect with a significant widening of the corresponding bands. Thus, probe field spectroscopy is also sufficiently sensitive to the considered effects and can serve as an alternative technique for studying the processes in material in the vicinity of the emitter. At the same time, the question of the specificity of the change in the spectral curves in the presence of several strong effects remains open. In order to illustrate this situation, let us consider the joint influence of ensemble local fields, the influence of the carrier as a continuous medium, and the influence of dephasing and widening.

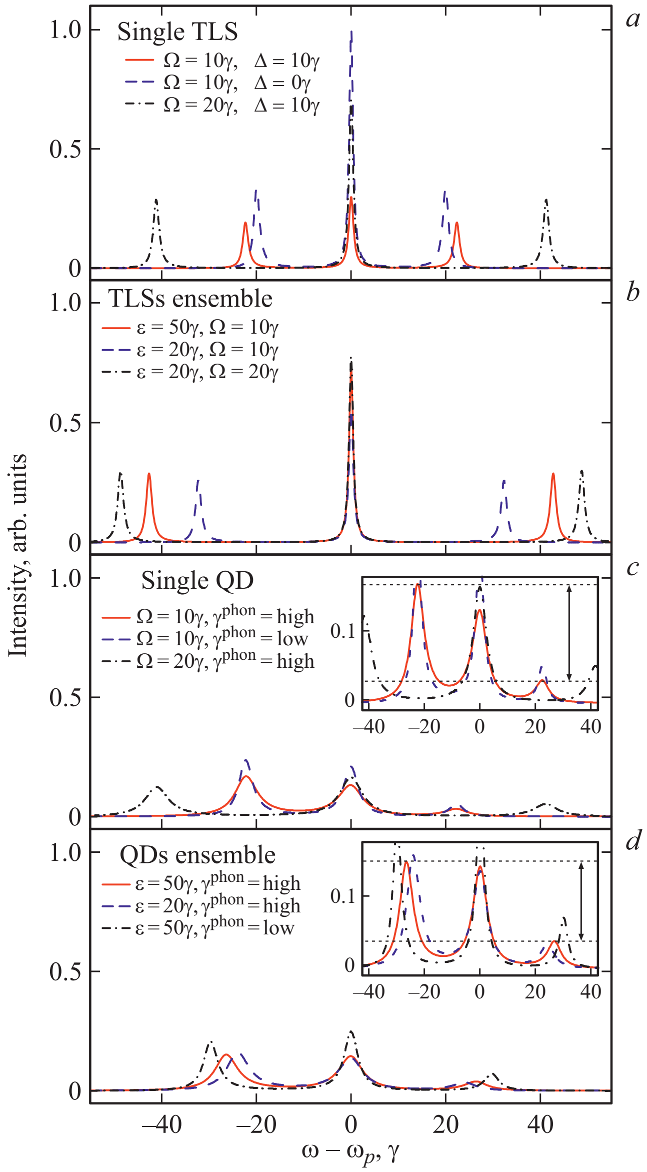

Figure 2. Fluorescence spectrum for various parameters: *a*) single two-level emitter; *b*) ensemble of two-level emitters (here, $\Delta = 10$, $\gamma_c = 0.9$), *c*) single model QD (here, $\Delta = 10$), where „$\gamma^{phon} = high$" means set of parameters $\gamma^+ = 0.9$, $\gamma^- = 0.8$, $\gamma^{cd} = 0.6$ and „$\gamma^{phon} = low$" stands for $\gamma^+ = 0.45$, $\gamma^- = 0.4$, $\gamma^{cd} = 0.3$; *d*) ensemble of model quantum dots (here, $\Omega = 10$, $\Delta = 10$, $\gamma_c = 0.9$).

To solve the latter problem, collective and dephasing effects should be taken into account simultaneously. This can be done formally by performing calculations for (13) and (16) using expressions (17) and (19). Figs. 1, *d* and 2, *d* show the corresponding results of calculations for the chosen parameters, allowing comparison with the previous spectral patterns. Noticeable mutual compensation of the effects is observed in the absorption spectra. The absorption and amplification bands are shifted, but occupy positions different from those for independent emitters. In this case, as can be seen, the balance of absorption and amplification can change depending on the ratio of the parameters of „competing" processes. A similar situation is observed in the resonance fluorescence spectrum. The spectral peaks are shifted relative to the positions determined by the frequency Ω, but the height ratio changes. In this case, the asymmetry with respect to both the heights and widths of the spectral lines is preserved.

4. Discussion and conclusion

The paper presents a theoretical analysis of the absorption spectra of the probe field and the resonance fluorescence spectra of two-level quantum emitters under various model conditions, when the absorption and emission of light depend on the parameters and state of the environment. A mathematical apparatus has been developed for calculating the particle emission spectra in the presence of a strong external field in the semiclassical approximation based on the well-known formalism, i.e. the probe field method. The emission spectrum can be restored from the analysis of the absorption/amplification factor of weak probe signal with wavelength tuning in the vicinity of the transition of the emitter interacting with the control monochromatic signal. The equivalence of the presented method with the quantum-kinetic approach based on Bogolyubov chains for the emitter density matrices and the field is shown. Calculations are made of the absorption spectra of weak probe signal propagating through a medium controlled by strong cw laser tuned near a single transition of optically active impurity centers. Resonance fluorescence spectra for models where light-emitting systems are affected by local fields and effects of local electron-phonon interactions are calculated.

It follows from the calculation results that the considered types of spectral dependences can be used to restore the characteristics of media containing probe particles —

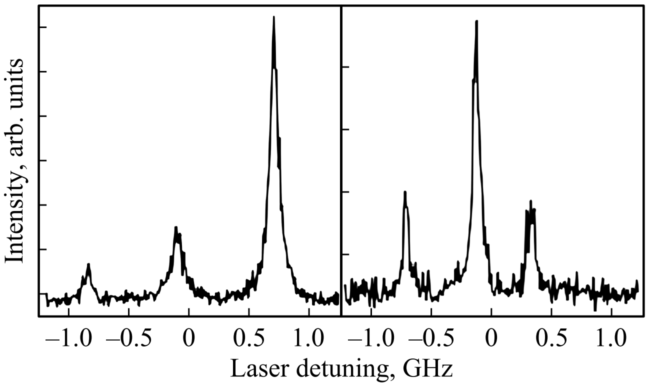

Figure 3. Photoluminescence excitation spectra of terylene molecules in a thin film of 1,2-ortho-dichlorobenzene.

emitters. The paper analyzes the phenomenon of resonant fluorescence of two-level particles. When this type of photoluminescence is realized, the spectra under study at sufficiently strong pumping have more than one frequency band, the characteristics of which may contain information about the state of the emitter and its participation in the processes in the medium near it. Thus, the mutual configuration of the parameters of each of the absorption and amplification contours of the probe field (frequency position, height, width) is determined by the pumping parameters, the presence of radiative corrections due to the medium and interaction with phonon modes. The analysis of the spectral contour dependence on the controlled pumping parameters (wavelength, power) and the state of the medium (temperature) makes it possible to determine the nature and types of processes accompanying photoluminescence. Similar possibilities can be provided by the analysis of Apanasevich−Mollow resonance fluorescence triplets. The frequency positions, heights and widths of the radiation peaks are also determined by the processes taking place in the medium around the emitter. Separate processes manifest themselves unambiguously against the background of controlled pumping and can be identified. The presence of a larger number of parameters (determining the state of a single transition) than in the analysis of a single emission line in certain cases can provide a more accurate restoration of the medium characteristics. The latter should be ensured by the development of theoretical knowledge about the optics of impurity particles.

In conclusion, note that the multivariative analysis of spectral triplets is not limited to the considered examples and, in particular, to the possibilities of using two-level emitters described in the paper. As mentioned above, pairs of two-level emitters can be introduced in solid medium and experience dipole-dipole entanglement. The photoluminescence excitation spectra (the total luminescence intensity dependence on the pumping frequency) of cooperative pairs also have the form of triplet. Examples of original excitation spectra of organic molecules are shown in Fig. 3. The Figures show that the excitation contours have the same distances between the peaks, but the peaks differ in height and width. It follows from this that the function of the resonance fluorescence triplet contour is not specific to systems of two-level emitters. At the same time, the sets of characteristics of the spectral maxima in the emission and excitation spectra are determined by different physical processes, but all are subject to the influence of the local environment.

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Conflict of interest

The authors declare that they have no conflict of interest.

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