

Approach for describing spatial dynamics of quantum light-matter interaction in dispersive dissipative media

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Solving the challenging problem of the amplification and generation of an electromagnetic field in nanostructures enables us to implement many properties of the electromagnetic field at the nanoscale in practical applications. A first-principles quantum-mechanical consideration of such a problem is sufficiently restricted by the exponentially large number of degrees of freedom and does not allow the electromagnetic-field dynamics to be described if it involves a high number of interacting atoms and modes of the electromagnetic field. Conversely, the classical description of electromagnetic fields is incorrect at the nanoscale due to the high level of quantum fluctuations connected to high dissipation and noise levels. In this paper, we develop a framework with a significantly reduced number of degrees of freedom, which describes the quantum spatial dynamics of electromagnetic fields interacting with atoms. As an example, we consider the interaction between atoms placed in a metallic subwavelength groove and demonstrate that a spontaneously excited electromagnetic pulse propagates with the group velocity. The developed approach may be exploited to describe nonuniform amplification and propagation of electromagnetic fields in arbitrary dispersive dissipative systems.

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I. INTRODUCTION

The study of the interaction between light and matter is a key problem in physics [1]. Progress in nanotechnologies [2–5] has made it possible to enhance light-matter interaction at the nanoscale. Such an enhancement plays a crucial role in investigating the influence of the electromagnetic environment, such as photonic crystals and metallic and dielectric plasmonic structures, on the atomic dynamics [6,7]. In such structures, engineering of the electromagnetic-field density of states allows light-matter interaction to be controlled [8]. This control enables conditions for excitation and coherent generation of the electromagnetic field to be achieved. This allows devices like distributed feedback (DFB) lasers [9–21], nanolasers, and spasers [22–35] to be created.

Consistent consideration of the dynamics of electromagnetic fields and atoms is based on quantum electrodynamics. The quantum properties of light arise in theory after the procedure of field quantization, which implies that the electromagnetic field is expanded in a series of system eigenmodes [6,36–38]. With excitation of the mode, the electromagnetic field appears in the entire mode volume. Thus, if it is essential to consider the temporal evolution of the electromagnetic field in a finite volume, then it is necessary to take into account the infinite number of modes with an appropriate phase relation [6,36–38].

Accurate description of the temporal evolution of electromagnetic field is necessary in the problem of map coherent superposition from one quantum bit to another by means of an electromagnetic field [39–42], the study of ultrafast active plasmonics [34,43–45], the study of laser dynamics with pulsed pumping [11,16], and lasers with an inhomogeneous

distribution of gain medium or cavity-free lasers [30,35,46,47]. For example, in the cavity-free lasers the generation threshold depends on the time an electromagnetic pulse emitted by an active atom stays inside the active region and interacts with other active atoms. This time depends on the group velocity. In nanostructures it may be four orders of magnitude smaller than the speed of light [11,46]. This makes the time of mode propagation along the structure comparable with characteristic nanolaser times [48]. Thus, the effects of retardation should be taken into account when considering operations of nanolaser devices.

In a full quantum-mechanical consideration, the increase in the number of modes leads to the exponential increase in the number of degrees of freedom [49]. The same increase takes place when the number of atoms increases. As a result, a first-principles consideration of the problem of the interaction between atoms and modes of electromagnetic field is impossible for many practical applications. It should be underlined that even in the simplest cases, the first-principles quantum-mechanical consideration of spatial dynamics is complicated. For example, the problem of the finiteness of the propagation speed of an electromagnetic signal between two atoms (the so-called Fermi problem [50]) has a long history and was solved only recently [51–59].

Effects related to the quantum nature of the electromagnetic field and atoms in many practical problems can be addressed without involving exact quantum-mechanical calculations. There are mean-field theories describing the dynamics of a finite number of physical values, neglecting quantum-mechanical correlations [60–62]. Among these theories, the most extensively used are the rate equations and the Maxwell-Bloch equations [60–62]. The rate equations can be implemented for laser description, accounting for the spontaneous decay of atoms [60–62]. They are in good agreement with the experimental data for lasers with high-quality cavities at

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large time scales when the stationary regime is set. However, the rate equations do not take into account the phase relations between electromagnetic waves; thus, they are not appropriate for describing many important effects in modern physics, e.g., the propagation of an electromagnetic pulse [60]. In the Maxwell-Bloch equations, the classical description of an electromagnetic field is used [60–62]. Although this approach uses the wave equation for the field and the finiteness of the propagation speed of the electromagnetic field, it does not take into account the spontaneous decay of atoms [60]. To describe the process of the spontaneous decay, the noise operators are added to the Maxwell-Bloch equations [63–67]. For large numbers of photons and atoms that collectively participate in coherent and fluctuation dynamics, operator equations may be translated into c -number equations [64]. However, even in this case, the numerical simulation of these equations requires large computational resources [34,68,69]. Moreover, this approach is not suitable for nanosize systems, where the number of photons in the cavity and atoms is small [64].

The aim of this work is to develop a method to describe atom and electromagnetic mode interaction, taking into account the process of spontaneous emission and the finiteness of the propagation speed of electromagnetic fields in dispersive dissipative media. For this purpose, we obtained an equation system in which the number of equations is a quadratic function of the number of modes and a linear function of the number of atoms. In the case of a full quantum-mechanical calculation through master equations for the density matrix, the number of equations increases exponentially with the number of atoms or modes [49]. Implementing our approach makes the consideration of quantum systems with a large number of interacting atoms and modes possible. It is shown that the developed formalism correctly describes the propagation of an electromagnetic pulse with the group velocity. We show that the finiteness of the pulse propagation arises from interference between the electromagnetic fields of the different modes. We demonstrate that the rate equations describe interference erroneously and do not correctly describe ultrafast dynamics. We show that the electromagnetic pulse that is spontaneously emitted by an atom takes the form of a δ function at the initial time because the rate of spontaneous emission in different cavity modes at the initial time does not depend on the difference between atom transition frequency and the eigenfrequency of the mode. We also investigate the influence of the retardation effects on the multimode lasing and show that they substantially modify the laser threshold.

II. DESCRIPTION OF INTERACTION BETWEEN LIGHT AND MATTER

The dynamics of an interacting electromagnetic field and atoms in a Markov approximation is described by the master equation in the Lindblad form [49,63]:

$$\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar}[\hat{H}_a + \hat{H}_\sigma + \hat{V}, \hat{\rho}] + \hat{L}_a[\hat{\rho}] + \hat{L}_\sigma^e[\hat{\rho}] + \hat{L}_\sigma^{ph}[\hat{\rho}] + \hat{L}_\sigma^{\text{pump}}[\hat{\rho}], \quad (1)$$

where $\hat{H}_a = \sum_j \hbar \omega_j^a \hat{a}_j^\dagger \hat{a}_j$ is the Hamiltonian of the electromagnetic field after mode decomposition, $\hat{H}_\sigma =$

$\sum_m \hbar \omega_m^\sigma \hat{\sigma}_m^\dagger \hat{\sigma}_m$ is the Hamiltonian of the two-level atoms, and $\hat{V} = \sum_{j,m} (\hbar \Omega_{jm} \hat{a}_j^\dagger \hat{\sigma}_m + \hbar \Omega_{jm}^* \hat{a}_j \hat{\sigma}_m^\dagger)$ is the interaction between modes and atoms in the Jaynes-Cummings form. Here $\hat{a}_j^\dagger$ and $\hat{a}_j$ are, respectively, the creation and annihilation operators of photons in the j th mode, $\hat{\sigma}_m^\dagger$ and $\hat{\sigma}_m$ are, respectively, the raising and lowering operators for the transition of the m th two-level atom, Ω_{jm} is a coupling constant between the photons in the j th cavity mode and the m th atom, ω_j^a is an eigenfrequency of the j th cavity mode, and, finally, ω_m^σ is the transient frequency of the m th atom. The term $\hat{L}_a[\hat{\rho}] = \sum_j \frac{\gamma_j^a}{2} (2\hat{a}_j \hat{\rho} \hat{a}_j^\dagger - \hat{a}_j^\dagger \hat{a}_j \hat{\rho} - \hat{\rho} \hat{a}_j^\dagger \hat{a}_j)$ [49,63] describes energy relaxation in each j th mode with rate γ_j^a , $\hat{L}_\sigma^e[\hat{\rho}] = \sum_m \frac{\gamma_m^e}{2} (2\hat{\sigma}_m \hat{\rho} \hat{\sigma}_m^\dagger - \hat{\sigma}_m^\dagger \hat{\sigma}_m \hat{\rho} - \hat{\rho} \hat{\sigma}_m^\dagger \hat{\sigma}_m)$, and $\hat{L}_\sigma^{ph}[\hat{\rho}] = \sum_m \frac{\gamma_m^{ph}}{4} (\hat{D}_m \hat{\rho} \hat{D}_m - \hat{\rho})$, corresponding to the energy and phase relaxations in each atom with the rates γ_m^D and γ_m^{ph} , respectively [49,63]; the term $\hat{L}_\sigma^{\text{pump}}[\hat{\rho}] = \sum_m \frac{\gamma_m^{\text{pump}}}{2} (2\hat{\sigma}_m^\dagger \hat{\rho} \hat{\sigma}_m - \hat{\sigma}_m \hat{\sigma}_m^\dagger \hat{\rho} - \hat{\rho} \hat{\sigma}_m \hat{\sigma}_m^\dagger)$ describes pumping of a two-level atom with the rate γ_m^{pump} [49,63].

The Markov approximation is applicable when the time scale of the reservoir degrees of freedom is much shorter than the relaxation time of the system [49]. The characteristic correlation time of a reservoir which is in thermal equilibrium may be estimated as $\hbar/kT$ [49] and at room temperature is of the order of 10^{-14} s. To determine the applicability of the Markov approximation we need to compare this correlation time with the characteristic relaxation time of the system. For an active medium the relaxation time of the energy is 10^{-11} – 10^{-9} s; the time of the phase relaxation is 10^{-13} s. For nanocavity modes the relaxation time is 10^{-13} – 10^{-11} s. So we see that for both resonator modes and an active medium the relaxation time is at least one order of magnitude larger than the correlation time of the reservoir; that is, the Markovian approximation is applicable. An overwhelming majority of experiments with a metallic nanocavity confirm the Markovian model for describing the losses showing exponential decay [70].

Using the identity $\langle \hat{A} \rangle = \text{Tr}(\hat{\rho} \hat{A})$ and master equation (1), it is possible to derive a closed system of equations on operator average $D_m = \langle \hat{\sigma}_m^\dagger \hat{\sigma}_m - \hat{\sigma}_m \hat{\sigma}_m^\dagger \rangle$, $\varphi_{jm} = \langle -i \hat{a}_j^\dagger \hat{\sigma}_m \rangle$, and $n_{jl} = \langle \hat{a}_j^\dagger \hat{a}_l \rangle$. To this end, we make two approximations. The first one is the splitting correlations between the average values of the number of photons and the population inversion, $\langle \hat{n}_{jl} \hat{D}_m \rangle = \langle \hat{n}_{jl} \rangle \langle \hat{D}_m \rangle$. It is valid when the number of atoms $N_a \gg 1$ [49], which takes place in nanolaser experiments [48]. Under this condition all operators behave like c -numbers. Such uncoupling is used in the derivation of rate equations to take into account spontaneous emission [61,71]. The second approximation neglects quantum-mechanical correlations between different atoms, i.e., $\langle \hat{\sigma}_m^\dagger \hat{\sigma}_{m'} \rangle = \delta_{mm'} (\langle \hat{D}_m \rangle + 1)/2$ [60]. Such correlations are important in a system of entangled ions [72], superconducting qubits [73], Bose-Einstein condensates [74,75], and light sources based on alkaline-earth atoms [76,77]. The rate of destroying such correlations increases with the temperature and the number of atoms [72,78]. Such correlations appear only at temperatures of the order of K and a number of atoms of the order of 10 [72,78]. Such correlations are not exhibited in nanolasers, which usually operate at room temperature and at a high number of atoms of the active medium, and below we will neglect them.

This results in the following equations:

$$\frac{dn_{jl}}{dt} = -\gamma_{jl}^n n_{jl} + i(\omega_j^a - \omega_l^a) n_{jl} + \sum_m (\Omega_{lm} \varphi_{jm} + \Omega_{jm}^* \varphi_{lm}^*), \quad (2)$$

$$\frac{dD_m}{dt} = -\gamma_m^D (1 + D_m) + \gamma_m^{\text{pump}} (1 - D_m) - 2 \sum_j (\Omega_{jm} \varphi_{jm} + \Omega_{jm}^* \varphi_{jm}^*), \quad (3)$$

$$\frac{d\varphi_{jm}}{dt} = -\gamma_{jm}^\varphi \varphi_{jm} + i(\omega_j^a - \omega_m^\sigma) \varphi_{jm} + \frac{\Omega_{jm}^*}{2} (D_m + 1) + \sum_l \Omega_{lm}^* n_{jl} D_m. \quad (4)$$

In Eqs. (2)–(4), D_m is the average value of the operator of the population inversion of the m th atom [61,62], while $\varphi_{jm} = \langle -i\hat{a}_j^\dagger \hat{\sigma}_m \rangle$ is the average value of the operator that describes the interaction between the electromagnetic field in the j th cavity mode and the m th atom, $\gamma_{jm}^\varphi = (\gamma_j^a + \gamma_m^D + \gamma_m^{\text{pump}} + \gamma_m^{ph})/2$. Here n_{jl} is the average value of the operator of the number of photons in the j th cavity mode when $j = l$, and n_{jl} is the average value of the operator that describes the transition of photons from the l th cavity mode to the j th cavity mode when $j \neq l$, $\gamma_{jl}^n = (\gamma_j^a + \gamma_l^a)/2$. This operator arises from the interference between the electromagnetic field in the j th and l th cavity modes. Below, we will demonstrate that neglecting these terms (i.e., interference between the electromagnetic fields of different cavity modes) results in instant propagation of the electromagnetic field. We will name the variables n_{jl} cross terms.

Note that the rate equations may be obtained from Eqs. (2)–(4) [60]. First, neglecting the phase relations between electromagnetic modes $\langle \hat{a}_j^\dagger \hat{a}_l \rangle = \delta_{jl} \langle \hat{n}_{jj} \rangle$, it is possible to reduce Eqs. (2)–(4) to

$$\frac{dn_j}{dt} = -\gamma_j^a n_j + \sum_m (\Omega_{jm} \varphi_{jm} + \Omega_{jm}^* \varphi_{jm}^*), \quad (5)$$

$$\frac{dD_m}{dt} = -\gamma_m^D (1 + D_m) + \gamma_m^{\text{pump}} (1 - D_m) - 2 \sum_j (\Omega_{jm} \varphi_{jm} + \Omega_{jm}^* \varphi_{jm}^*), \quad (6)$$

$$\frac{d\varphi_{jm}}{dt} = -\gamma_{jm}^\varphi \varphi_{jm} + i(\omega_j^a - \omega_m^\sigma) \varphi_{jm} + \frac{\Omega_{jm}^*}{2} (D_m + 1) + \Omega_{jm}^* n_j D_m, \quad (7)$$

where $n_j = n_{jj}$ is the average value of the number of photons in the j th cavity mode. These equations have been used to describe the emission properties of micro- and nanolasers [79–81].

Second, in most types of lasers, the rate of transverse relaxation is larger than that of longitudinal relaxation and the decay rate of the number of photons (i.e., $\gamma_m^{ph} \gg \gamma_m^D, \gamma_m^{\text{pump}}, \gamma_j^a$). In

this case, φ_{jm} can be adiabatically eliminated from Eqs. (5)–(7) [61], which results in

$$\frac{dn_j}{dt} = -\gamma_j^a n_j + \sum_m \frac{\gamma_{jm}^\varphi |\Omega_{jm}|^2}{(\gamma_{jm}^\varphi)^2 + (\omega_m^\sigma - \omega_j^a)^2} (2n_j D_m + D_m + 1), \quad (8)$$

$$\frac{dD_m}{dt} = -\gamma_m^D (1 + D_m) + \gamma_m^{\text{pump}} (1 - D_m) - 2 \sum_j \frac{\gamma_{jm}^\varphi |\Omega_{jm}|^2}{(\gamma_{jm}^\varphi)^2 + (\omega_m^\sigma - \omega_j^a)^2} (2n_j D_m + D_m + 1). \quad (9)$$

Equations (8) and (9) are the rate equations, also known as the balance equations. They can also be derived from the energy balance [61]. We show below that both Eqs. (5)–(7) and Eqs. (8) and (9) are incorrect for the description of the propagation of electromagnetic pulses.

III. PROPAGATION OF ELECTROMAGNETIC FIELD BETWEEN TWO ATOMS

To demonstrate the main features of Eqs. (2)–(4) and their advantages over the rate equations, we consider two two-level atoms placed in the subwavelength groove in metal [Fig. 1(a)]. In the case of a groove that is straight along the z axis with the profile function $y = \zeta(x)$, the eigenmodes of such systems may be presented in the form $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_{k\omega}(x, y) \exp(ikz - i\omega t)$, $\mathbf{H}(\mathbf{r}, t) = \mathbf{H}_{k\omega}(x, y) \exp(ikz - i\omega t)$, where $\mathbf{E}_{k\omega}(x, y)$ and $\mathbf{H}_{k\omega}(x, y)$ are determined through the following equation:

$$[\partial^2/\partial x^2 + \partial^2/\partial y^2 - \beta^2(k, \omega)] \begin{Bmatrix} \mathbf{E}_{k\omega}(x, y) \\ \mathbf{H}_{k\omega}(x, y) \end{Bmatrix} = 0, \quad (10)$$

where $\beta(k, \omega) = \sqrt{k^2 - \omega^2/c^2}$ in a vacuum and $\beta(k, \omega) = \sqrt{k^2 - \varepsilon(\omega)\omega^2/c^2}$ in the metal [explicit expressions for $\mathbf{E}_{k\omega}(x, y)$, $\mathbf{H}_{k\omega}(x, y)$, and the dispersion relation may be found in [82]].

After the quantization procedure, the electric and magnetic fields are expressed through creation and annihilation operators for each mode:

$$\hat{\mathbf{E}}(\mathbf{r}, t) = \sum_j A_0(k_j^a) \mathbf{E}_{k_j^a \omega_j^a}(x, y) \times \exp(ik_j^a z - i\omega_j^a t) \hat{a}_j + \text{H.c.}, \quad (11)$$

$$\hat{\mathbf{H}}(\mathbf{r}, t) = i \sum_j A_0(k_j^a) \mathbf{H}_{k_j^a \omega_j^a}(x, y) \times \exp(ik_j^a z - i\omega_j^a t) \hat{a}_j + \text{H.c.} \quad (12)$$

The operators $\hat{a}_j$ satisfy the boson commutation relations $[\hat{a}_j, \hat{a}_l^\dagger] = \delta_{jl} \hat{1}$; the dimensional constant $A_0(k_j^a)$ is determined

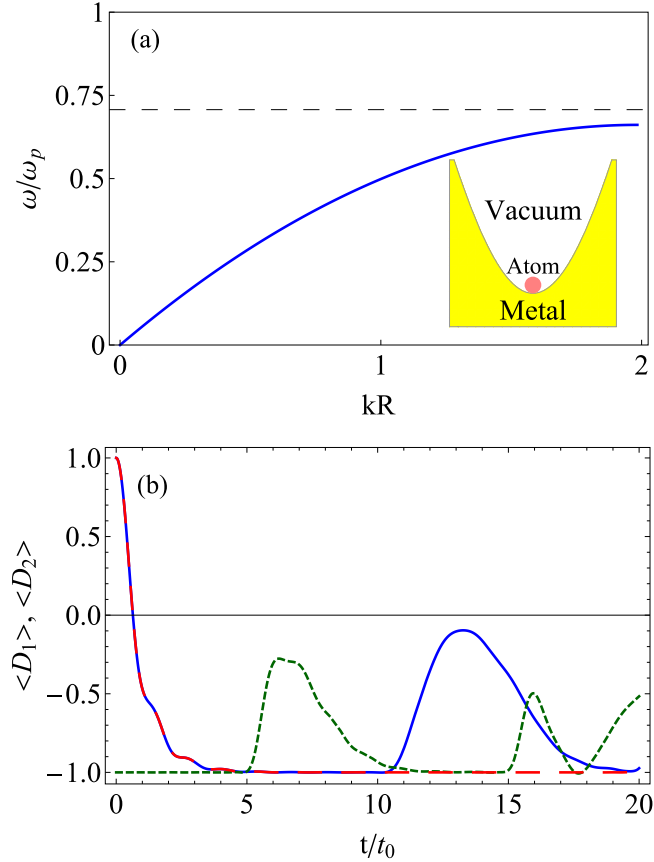


FIG. 1. (a) Dispersion curve of the eigenmodes of the metallic groove with the profile $\zeta(x) = -A \exp(-x^2/R^2)$. Inset: schematically illustrated metallic groove. (b) Dependence of population inversion of the first (blue solid line) and second (green dashed line) atoms and the dependence of the population inversion of the first atom with time in the absence of the second atom (red dot-dashed line) obtained from Eqs. (2)–(4). The distance between the atoms is $l = 500\lambda_\sigma$, the groove length is $L = 2000\lambda_\sigma$, and the atoms are placed symmetrically with respect to the groove center. Here t_0 is equal to $10^2\lambda_\sigma/v_g$, where λ_σ is the wavelength of the atom transition; $v_g = (\partial\omega/\partial k)|_{\omega=\omega_\sigma}$ is group velocity.

by the following condition [83–86]:

$$A_0^2(k_j^a) \frac{L}{8\pi} \int dx dy \left[\partial(\varepsilon\omega)/\partial\omega |\mathbf{E}_{k_j^a\omega_j^a}(x,y)|^2 + |\mathbf{H}_{k_j^a\omega_j^a}(x,y)|^2 \right] = \hbar\omega_j^a, \quad (13)$$

where L is the groove length.

The interaction between electromagnetic fields and atomic dipole moments in the rotating-wave approximation takes the form of the Jaynes-Cummings Hamiltonian:

$$\hat{V} = \sum_{j,m} (\hbar\Omega_{jm}\hat{a}_j^\dagger\hat{\sigma}_m + \hbar\Omega_{jm}^*\hat{a}_j\hat{\sigma}_m^\dagger), \quad (14)$$

with the Rabi frequency

$$\begin{aligned} \Omega_{jm} &= -\mathbf{d}_{eg}^m A_0(k_j^a) \mathbf{E}_{k_j^a\omega_j^a}(x_m, y_m) \exp(ik_j^a z_m)/\hbar \\ &= \Omega_{j0} \exp(ik_j^a z_m), \end{aligned} \quad (15)$$

where $\mathbf{r}_m = \{x_m, y_m, z_m\}$ and $\mathbf{d}_{eg}^m$ are coordinate and matrix elements of the dipole transition of the m th atom, respectively. In the last equality we suppose that each atom has the same lateral coordinates, $\{x_m, y_m\} = \{x, y\}$.

The decay rate of each mode with wave vector k_j^a is determined by the part of electromagnetic energy inside the metal and may be evaluated as [87,88]

$$\gamma_j^a = \frac{\omega_j^a \int_{\text{metal}} dx dy (\varepsilon'' |\mathbf{E}_{k_j^a\omega_j^a}(x,y)|^2)}{\int dx dy \left(\frac{\partial(\varepsilon\omega)}{\partial\omega} |\mathbf{E}_{k_j^a\omega_j^a}(x,y)|^2 + |\mathbf{H}_{k_j^a\omega_j^a}(x,y)|^2 \right)}. \quad (16)$$

Let us consider a system of two identical atoms with $\omega_\sigma^m = \omega_\sigma = 0.6\omega_p$ (ω_p is the plasma frequency of metal), $\gamma_m^D = 10^{-6}\omega_\sigma$, and $\gamma_m^{ph} = 10^{-2}\omega_\sigma$ that are placed at a distance of l from each another in the groove with the profile $\zeta(x) = -A \exp(-x^2/R^2)$, whose dispersion curve is calculated in [82] [see Fig. 1(a)]. The first atom at the initial time is in the excited state [i.e., $D_1(0) = 1$]; the second atom at the initial time is in the ground state [i.e., $D_2(0) = -1$], and the number of photons in the cavity is equal to zero. As the operators $\hat{n}_{jl}$ ($j \neq l$) and $\hat{\varphi}_{jm}$ change the number of photons in the cavity modes, the average values of these operators n_{jl} and φ_{jm} are equal to zero when the system is in the Fock state (e.g., when the number of photons is equal to zero).

Using Eqs. (2)–(4), we calculated the dependence of the population inversion of the first and second atoms with time [Fig. 1(b)]. As follows from Eqs. (2)–(4), the population inversion of the first atom starts to decay at the initial time [Fig. 1(b)]. The population inversion of the second atom is constant and equal to -1 until the time is less than propagation time τ_{pr} . After the electromagnetic pulse has reached the second atom, its population inversion begins to increase. The presence of the second atom has no effect on the population inversion of the first atom, until the time is less than two propagation times $2\tau_{pr}$ [see the blue solid and red dot-dashed lines in Fig. 1(b)]. We investigate the dependence of the propagation time τ_{pr} on the distance between atoms and show that τ_{pr} is a linear function of the distance between atoms [Fig. 2(a)]. This means that the electromagnetic pulse propagates with constant velocity. To investigate the physical nature of this velocity, we change the transition frequency ω_σ of the atoms. This leads to a change in the group and phase velocities of the spontaneously emitted electromagnetic pulse at the frequency ω_σ [Fig. 2(b), blue solid and dashed lines]. The numerical simulation of Eqs. (2)–(4) shows that the population inversion of the second atom begins to increase after the time $\tau_{pr} = l/v_g$, where $v_g = (\partial\omega/\partial k)|_{\omega=\omega_\sigma}$ is the group velocity of the electromagnetic (EM) pulse for the system under consideration [see the coincidence of $\tau_{pr} = l/v_g$ (blue line) and the time of atom excitation (the bright region) in Fig. 2(b)]. Note that there is an apparent difference between $\tau_{pr} = l/v_g$ (blue line) and the time of atom excitation (the bright region) at the bottom of Fig. 2(b). The reason is the rate of increase of the population inversion is determined by the constant of interaction with the EM pulse, which is proportional to the density of states at the atomic transition frequency ω_σ . Because the density of states is inversely proportional to the group velocity [89,90], when the latter is small, the atom population inversion reaches its maximum value earlier. We emphasize

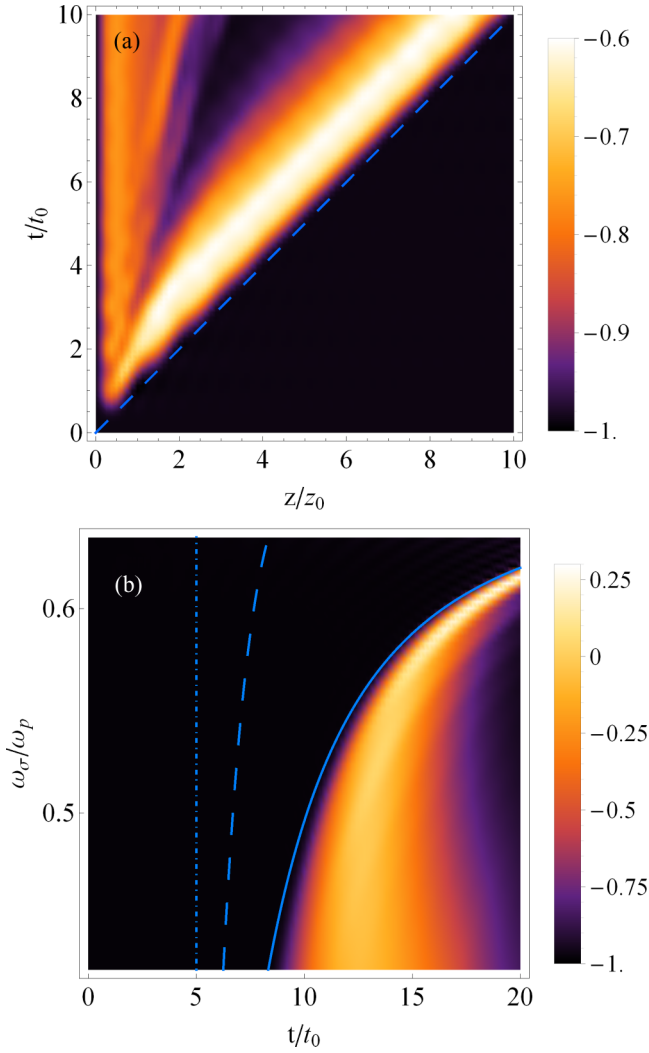


FIG. 2. (a) Dependence of the population inversion of the second atom on time and distance between atoms obtained from Eqs. (2)–(4). The blue dashed line is determined by group velocity $z = v_g t$. Here z_0 is equal to $10^2 \lambda_\sigma$, and t_0 is equal to $10^2 \lambda_\sigma / v_g$. (b) Dependence of the population inversion of the second atom on time and the transition frequency of the atoms obtained from Eqs. (2)–(4). The solid line is the curve $t = l/v_g$ [$v_g = (\partial\omega/\partial k)|_{\omega=\omega_\sigma}$ is group velocity], the dashed line is the curve $t = l/v_\phi$ [$v_\phi = (\omega/k)|_{\omega=\omega_\sigma}$ is phase velocity], and the dashed-dot line is the curve $t = l/c$.

that neither the phase velocity nor the speed of light in a vacuum affects the atomic population inversion.

Thus, Eqs. (2)–(4) take into account the process of spontaneous emission and the finiteness of the propagation speed of electromagnetic waves. The propagation speed of the electromagnetic signal is equal to the group velocity on the transition frequency of the atom.

IV. EQUATIONS WITHOUT CROSS TERMS

If we assume that $\langle \hat{a}_j^\dagger \hat{a}_l \rangle = \delta_{jl} \langle \hat{n}_{jj} \rangle$, then Eqs. (2)–(4) reduce to the equation system (5)–(7). When Eqs. (5)–(7) are implemented, the population inversion of the second atom begins to increase with no delay (Fig. 3), and the dynamics of

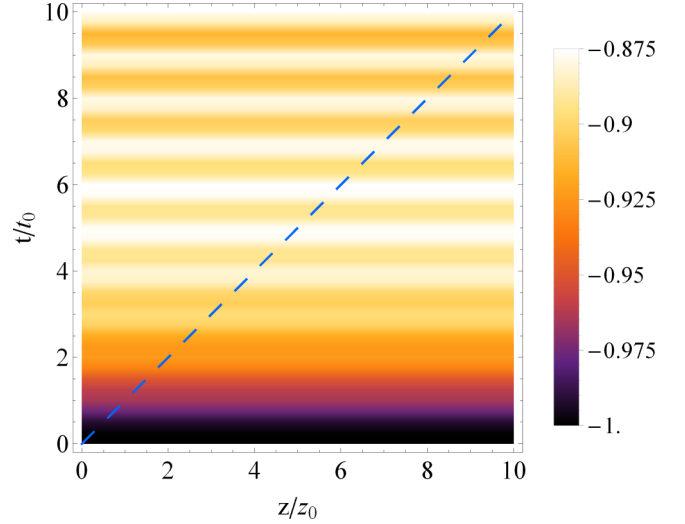


FIG. 3. Dependence of the population inversion of the second atom on time and distance between atoms obtained from Eqs. (5)–(7). The blue dashed line is determined by group velocity $z = v_g t$. Here z_0 is equal to $10^2 \lambda_\sigma$ where λ_σ is the wavelength of the atom radiation; $v_g = (\partial\omega/\partial k)|_{\omega=\omega_\sigma}$ is group velocity.

the population inversions of both atoms do not depend on the distance between them.

The reason for the incorrect dynamics description from Eqs. (5)–(7) can be explained by means of classical electrodynamics. The variables $\Omega_{jm} \varphi_{jm}$ describe the interaction between the electromagnetic field and the atoms [see Eq. (6)]. They are proportional to the intensity I of the electromagnetic field in the atom location, which is proportional to the square of the magnitude of the electric field. The electric field can be expanded in the following Fourier series:

$$E(z, t) = \sum_j [a_j \exp(ik_j^a z) + a_j^* \exp(-ik_j^a z)] \times \exp(-i\omega_j^a t). \quad (17)$$

The amplitudes of the Fourier harmonics a_j and a_j^* are classical analogs of the annihilation and creation operators $\hat{a}_j$ and $\hat{a}_j^\dagger$. The intensity of the electromagnetic field is written as

$$I(z) = \sum_j \sum_l \{a_j a_l^* \exp[i(k_j^a - k_l^a)z] + a_j^* a_l \exp[-i(k_j^a - k_l^a)z]\} \exp[-i(\omega_j^a - \omega_l^a)t] + \sum_j \sum_l \{a_j a_l \exp[i(k_j^a + k_l^a)z] + a_j^* a_l^* \exp[-i(k_j^a + k_l^a)z]\} \exp[-i(\omega_j^a + \omega_l^a)t]. \quad (18)$$

The terms in the first sum of Eq. (18) are the classical analogs of operators of $\hat{n}_{jl} = \hat{a}_j^\dagger \hat{a}_l$. Elements of the sum with $j \neq l$ describe classical mode interference. The terms in the second sum of Eq. (18) oscillate with double frequency. In the rotating-wave approximation, these terms are neglected.

Neglecting the cross terms ($\langle \hat{a}_j^\dagger \hat{a}_l \rangle = \delta_{jl} \langle \hat{n}_{jj} \rangle$) in Eqs. (2)–(4) is the equivalent of neglecting the interference terms

because Eq. (18) takes the following form:

$$\tilde{I}(z) = 2 \sum_j |a_j|^2. \quad (19)$$

The variable $\tilde{I}(z)$ does not depend on the coordinate, which leads to independence of the atomic interaction from the distance between atoms. This behavior is observed when we use Eqs. (5)–(7) to describe atomic interaction (Fig. 3).

Thus, neglecting the cross terms translates Eqs. (2)–(4) into Eqs. (5)–(7) and results in instant propagation of the electromagnetic field. As a result, Eqs. (5)–(7) do not allow for the finiteness of the propagation speed of the electromagnetic field. The rate equations (8) and (9) are derived from Eqs. (5)–(7) by the adiabatic elimination of the variables of φ_{jm} . Therefore, the rate equations do not take into account the finiteness of the propagation speed of the electromagnetic field.

V. FORM OF THE ELECTROMAGNETIC PULSE AT THE INITIAL TIME OF SPONTANEOUS DECAY

Based on an analogy with classical electrodynamics, we determine the following variable:

$$I(z, t) = \sum_j \sum_l [\Omega_l^*(z) \Omega_j(z) n_{jl}(t) + \Omega_l(z) \Omega_j^*(z) n_{jl}^*(t)], \quad (20)$$

which is proportional to the intensity of the electromagnetic field at the point z . Here we used the notation

$$\Omega_j(z) = \Omega_{j0} \exp(ik_j^a z), \quad (21)$$

which is similar to determining the coupling constant between the photons in the cavity modes and the atoms Ω_{jm} [see Eq. (15)].

As follows from Eqs. (2)–(4), the electromagnetic pulse that was emitted by the first atom propagates with the group velocity of the EM field and, at the initial time, has the form of a δ function (i.e., the electromagnetic field is different from zero only at the location of the first atom; Fig. 4).

The Fourier harmonics of the δ function are equal to one another:

$$\delta(z) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \exp(ikz) dk. \quad (22)$$

Thus, at the initial time of spontaneous decay, the electromagnetic pulse has Fourier harmonics equal to one another. This may be achieved only if all rates of spontaneous decay Γ_{sp} in every cavity mode are equal and independent of their eigenfrequencies.

In Eqs. (2)–(4), the rate of spontaneous decay of the m th atom in the j th cavity mode is proportional to the variable $\Omega_{jm} \varphi_{jm}$ [see Eq. (6)]. Time integration of Eq. (4) results in

$$\begin{aligned} \varphi_{jm}(t) = & [i(\omega_j^a - \omega_\sigma) - \gamma_{jm}^\varphi] \int_0^t \varphi_{jm} d\tau \\ & + \frac{\Omega_{jm}^*}{2} \int_0^t (D_m + 1) d\tau + \sum_l \Omega_{lm}^* \int_0^t n_{jl} D_m d\tau. \end{aligned} \quad (23)$$

At the beginning of the spontaneous decay, the number of photons is equal to zero. As the operators $\hat{n}_{jl}$ ($j \neq l$) and

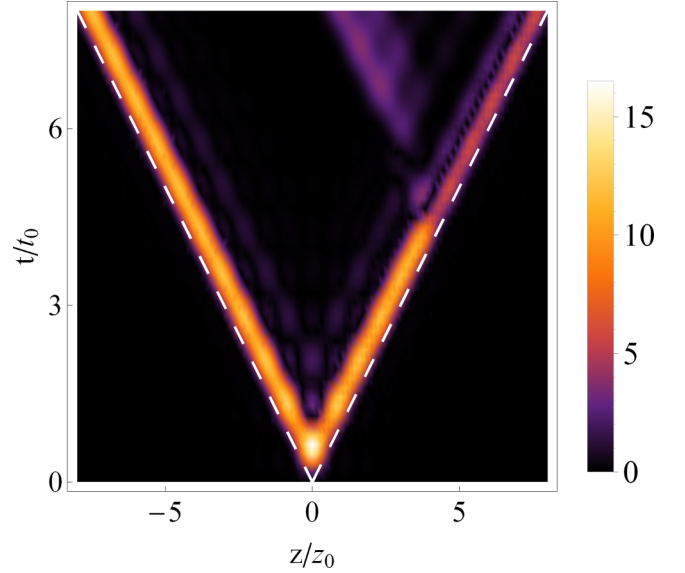


FIG. 4. Dependence of the intensity of the electromagnetic field in the cavity on time and coordinates obtained from Eqs. (2)–(4). The white dashed lines are determined by group velocity $z = v_g t$. The first atom is located at $z = 0$, and the second atom is located at $z = 400 \lambda_\sigma$. Here z_0 is equal to $10^2 \lambda_\sigma$, and t_0 is equal to $10^2 \lambda_\sigma / v_g$, where λ_σ is the wavelength of the atom radiation; $v_g = (\partial \omega / \partial k)|_{\omega=\omega_\sigma}$ is group velocity.

$\hat{\varphi}_{jm}$ change the number of photons in the cavity modes the average values of these operators n_{jl} and φ_{jm} are equal to zero when the system is in the Fock state (e.g., when the number of photons is equal to zero). As a result,

$$\int_0^t \varphi_{jm} d\tau \approx 0 \quad (24)$$

when the decay time t is less than the characteristic time of the problem.

Therefore, at the initial time, the rate of spontaneous decay does not depend on the difference between the eigenfrequency of the cavity mode and the frequency of the atom transition:

$$\begin{aligned} \Omega_{jm} \varphi_{jm}(t) & \approx \frac{|\Omega_{jm}|^2}{2} \int_0^t (D_m + 1) d\tau \\ & = \frac{|\Omega_{j0}|^2}{2} \int_0^t (D_m + 1) d\tau, \end{aligned} \quad (25)$$

where we used the determination of variables Ω_{jm} [see Eq. (15)]. As a result, the initial amplitudes of all cavity modes are equal to one another, and the electromagnetic pulse forms a δ function.

Note that in the rate equations (8) and (9), the rate of the spontaneous decay in the cavity mode is proportional to

$$\Gamma_{sp} \sim \frac{\gamma_{jm}^\varphi |\Omega_{jm}|^2}{(\gamma_{jm}^\varphi)^2 + (\omega_\sigma - \omega_j^a)^2}. \quad (26)$$

This factor depends on the difference between the eigenfrequency of the cavity mode and the frequency of the atom transition. Therefore, when the rate equations are used, the electromagnetic pulse does not form a δ function at the initial time. This is another reason why the rate equations

do not describe the finiteness of the propagation speed of an electromagnetic pulse.

VI. INFLUENCE OF CROSS TERMS ON MULTIMODE LASING

In this section we consider another example of the application of our approach. Namely, we investigate the generation of the plasmons in a parabolic waveguide with open ends. As the gain medium we consider 100 identical atoms which are placed symmetrically with respect to the center of the gold parabolic groove; the distance between atoms is $\delta l = 0.8\lambda_\sigma$ (all other parameters are the same as in Sec. III). Similar systems have been proposed as a nanosize source of coherent radiation [9,30,35,47,91,92] and for enhancing light-matter interaction in nanowaveguides [93–95].

Using Eqs. (2)–(4) we calculate the dependence of the electromagnetic-field intensity and the distribution on the pump intensity (see Fig. 5). It is shown that the intensity has an S-shaped dependence on the pumping. The electromagnetic-field distribution has a maximum at the edges of the gain

medium. The same behavior has been observed in recent experimental studies (see Figs. 2 and 4 in [35] and [9,30,47,91,92]). We leave more detailed comparison for future works.

It is interesting to note that the solution of the rate equations (8) and (9) does not exhibit the S-shaped dependence and show a uniform distribution of the electromagnetic field (see Fig. 5). The reason for this is that a spontaneously excited electromagnetic pulse instantly leaves the gain medium without interacting with other atoms. This results in a decrease in light-matter interaction and suppression of lasing [see Fig 5(a)]. This demonstrates the inapplicability of rate equations (8) and (9) for such laser systems and the necessity of taking into account cross terms.

At the same time the full quantum consideration of this system requires enormous computational resources. The dimension of the density matrix is $2^{2N_a} \times N^{2N_m}$ [49], where N_a and N_m are the numbers of atoms and modes, respectively, and N is the number of photons in each mode. Thus, if the system consists of 100 atoms and 30 modes and the maximum possible photon number in every mode is limited by 10, then the dimension of the density matrix is $(2^{100} \times 10^{30})^2 \approx 10^{120}$. This number coincides with the number of equations needed to be solved [49] so the full quantum description of the system is challenging. The approach developed in this paper based on Eqs. (2)–(4) requires solving $N_m \times (N_m + N_a) + N_a = 4000$ equations for the same system.

VII. CONCLUSIONS

We have developed a framework to treat the interaction of the electromagnetic field of arbitrary structures and atoms which takes into account the process of spontaneous decays and the finiteness of the propagation speed of the electromagnetic pulse. As a result, we derive Eqs. (2)–(4) with operator averages $D_m = \langle \hat{\sigma}_m^+ \hat{\sigma}_m - \hat{\sigma}_m \hat{\sigma}_m^+ \rangle$, $\varphi_{jm} = \langle -i\hat{a}_j^+ \hat{\sigma}_m \rangle$, and $n_{jl} = \langle \hat{a}_j^+ \hat{a}_l \rangle$, which allow us to describe the process of spontaneous decays and the finiteness of the propagation speed of the electromagnetic pulse. Unlike the master equations for the density matrix in which the number of equations increases exponentially with the number of atoms or modes - in our approach, the number of equations is a quadratic function of the number of modes and a linear function of the number of atoms. This opens the possibility to study open quantum systems consisting of a large number of interacting atoms and modes.

We have demonstrated that when our equations are used, the electromagnetic pulse propagates with the group velocity of the electromagnetic field and takes the form of a δ function at the initial time of spontaneous decay. It was shown that accounting for the cross terms $\langle \hat{n}_{jl} \rangle = \langle \hat{a}_j^+ \hat{a}_l \rangle$ is necessary for a valid description of the propagation of the electromagnetic pulse in space. Neglecting these terms results in instantaneous propagation of electromagnetic waves in space.

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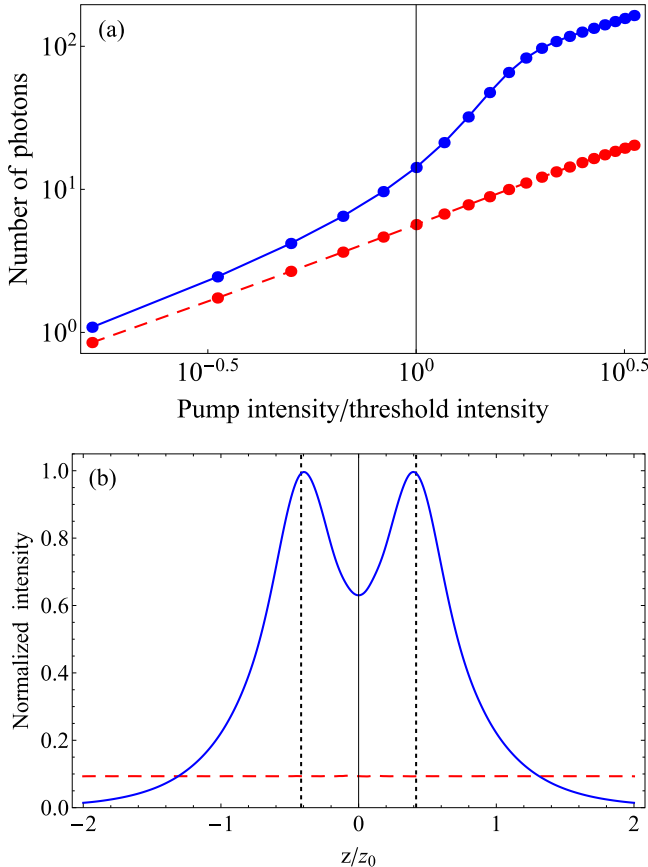


FIG. 5. (a) The dependence of the number of photons on the pumping obtained from Eqs. (2)–(4) (blue solid line) and from Eqs. (8) and (9) (red dashed line). The black vertical line corresponds to the lasing threshold. (b) The dependence of the normalized electromagnetic-field intensity on the coordinate obtained from Eqs. (2)–(4) (blue solid line) and from rate equations (8) and (9) (red dashed line). Vertical dashed lines correspond to the edges of the gain medium.

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