Light trapping in an ensemble of pointlike impurity centers in a Fabry-Perot cavity

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We report the development of quantum microscopic theory of quasiresonant dipole-dipole interaction in the ensembles of impurity atoms imbedded into transparent dielectric and located in a Fabry-Perot cavity. On the basis of the general approach we study the simultaneous influence of the cavity and resonant dipole-dipole interaction on the shape of the line of atomic transition as well as on light trapping in dense impurity ensembles. We analyze this influence depending on the size of the ensemble, its density, as well as on rms deviation of the transition frequency shifts caused by the symmetry disturbance of the internal fields of the dielectric medium. Obtained results are compared with the case when the cavity is absent. We show that the cavity can essentially modify cooperative polyatomic effects.

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1. INTRODUCTION

Ensembles of impurity centers (atoms) imbedded into a dielectric matrix are considered as promising objects for a wide range of problems in modern quantum optics and quantum electronics. The efficiency of their use, especially for optical applications, depends to a large measure on optical depth of the ensembles. Optical thickness can be increased by extension of the system and/or by decreasing of the mean free path of photons inside it.

In many cases increasing the size is undesirable or even impossible. In this situation the simplest and direct way to increase the optical depth is increasing the density of impurities. This way enhances the collective effects especially when the photon mean free path becomes comparable or less than resonant wavelength. In such a case resonant dipole-dipole interatomic interaction leads to density-dependent broadening and shifts of atomic transition as well as distortion of spectral line shape. Such collective effects were studied in detail in the case of nondegenerate cold atomic gases both experimentally [\[1–4\]](#page-8-0) (see also references therein) and theoretically, to name a few [\[5–](#page-8-0)[9\]](#page-9-0).

Besides resonant interatomic interaction the line shape of impurity centers is transformed due to interaction with the surrounding dielectric medium (see [\[10–20\]](#page-9-0)). Even in the case of a transparent dielectric the internal fields of a medium cause spectral line shifts which depends mainly on the type of chemical bond of a dielectric, the symmetry of the internal fields, and the temperature. These shifts can exceed the natural linewidth.

Another way to increase efficiency of light interaction with impurity ensembles is to use optical cavity or waveguide. A cavity offers an exciting tool to control over the lightmatter interaction. Since the seminal work of Purcell [\[21\]](#page-9-0) the peculiarities of atomic radiative properties, in particular, the enhancement and inhibition of the spontaneous decay rate inside a cavity or waveguide as well as near its surface has attracted a considerable attention [\[22–25\]](#page-9-0). Light matter interface in the presence of nanophotonic structures, such as nanofibers [\[26–28\]](#page-9-0), photonic crystal cavities [\[29\]](#page-9-0), and waveguides [\[30,31\]](#page-9-0) propose future applications for quantum metrology, scalable quantum networks, and quantum information science [\[32–34\]](#page-9-0).

Cavity modifies the structure of modes of electromagnetic field. It causes not only modification of spontaneous decay but also the nature of photon exchange between different atoms. In its turn it leads to alteration in dipole-dipole interatomic interaction [\[35\]](#page-9-0) as well as associated cooperative effects [\[36\]](#page-9-0). As opposed to spontaneous decay the modification of the dipole-dipole interaction is studied in less detail. By now there are several works dedicated to the interatomic interaction in the atomic systems coupled to a nanofiber [\[37\]](#page-9-0) as well as to phonic crystals [\[38–40\]](#page-9-0). Nevertheless, polyatomic cooperative effects inside the cavity, including multiple and recurrent scattering, have not been studied in detail yet.

The main goal of the present work is to analyze theoretically polyatomic cooperative effects in an ensemble of pointlike impurity centers embedded in a solid dielectric in the Fabry-Perot microcavity. We developed a consistent quantum theoretical approach based on approximate calculation of wave function of the joint system consisting of $N \gg 1$ motionless centers and the electromagnetic field. The interaction of impurity atoms with the dielectric is simulated by introduction inhomogeneous level shifts of the atomic energy levels.

As an example of a practical implementation of this approach in the present work we consider spontaneous decay of the local atomic excitation prepared inside a cavity. We calculate the transition spectrum of an excited atom and study radiation trapping in the considered system. We analyze the role of the cavity depending on the size of impurity ensemble, its density, as well as on rms deviation of the transition frequency shifts of the impurities caused by internal fields of the dielectric medium. Special attention is given to the case when the distance between two mirrors is less than a half of the transition wavelength. This case is of particular interest due to practically complete suppression of spontaneous decay of some Zeeman sublevels of atomic exited state. Obtained results are compared with the case when the cavity is absent.

2. BASIC ASSUMPTIONS AND APPROACH

Let us consider an ensemble, which consists of *N* motionless impurity atoms imbedded into transparent dielectric and placed in a Fabry-Perot cavity. The mirrors of a cavity are assumed to be perfectly conducting. The longitudinal sizes of

FIG. 1. Sketch of the cavity and the atomic ensemble.

the mirrors are much larger than resonant transition wavelength λ_0 , the distance between the mirrors *d* and the average distance between impurity atoms (see Fig. 1). It will allow us to consider the limit of infinite sizes of the mirrors in the final expressions.

We assume that the temperature is low enough to neglect the electron-phonon interaction. Influence of the dielectric on impurity atoms is simulated by means of random shift of their energetic levels. We think that the transition frequency of impurity atoms in a dielectric ω_a differs from the transition frequency of a free atom $\omega_0 - \omega_a = \omega_0 + \Delta_a$, where Δ_a is the frequency shift of the atom a ($a = 1,...,N$) which depends on its spatial position due to inhomogeneity of internal fields in dielectric.

All these assumptions allow us to consider dynamics of the model system which consists of the set of motionless pointlike scatterers and electromagnetic field in ideal cavity. We take into account all modes of the field including modes which are initially in the vacuum state. During the evolution these modes can be populated as a result of atomic decay, i.e., as a result of interaction of the excited atom with the field in the vacuum state. The photon created in such atomic transitions can be absorbed by another atom in the ensemble. This atom emits a secondary photon and so on. Thus, in our theory we deal with the closed quantum system, which can be described by the wave function.

In this paper we use the quantum microscopic approach described first in [\[41\]](#page-9-0) and developed afterward in [\[42\]](#page-9-0) for a description of collective effects in dense and cold nondegenerate atomic gases. This approach was earlier successfully used for analysis of optical properties of dense atomic ensembles [\[43–45\]](#page-9-0) as well as for studying light scattering from such ensembles [\[46–48\]](#page-9-0).

It is based on the solution of the nonstationary Schrodinger equation for the wave function ψ of the joint system consisting

of atoms and the electromagnetic field, including vacuum reservoir,

$$
i\hbar \frac{\partial \psi}{\partial t} = \widehat{H}\psi.
$$
 (2.1)

The Hamiltonian H of the joint system can be presented as a sum of Hamiltonian H_a of the atoms noninteracting with the field, the Hamiltonian H_f of the free field in a Fabry-Perot cavity, and the operator \hat{V} of its interaction,

$$
\widehat{H} = \widehat{H}_0 + \widehat{V},\tag{2.2}
$$

$$
\widehat{H}_0 = \widehat{H}_f + \sum_a \widehat{H}_a.
$$
\n(2.3)

In the dipole approximation used here, we have

$$
\widehat{V} = -\sum_{a} \widehat{\mathbf{d}}^{(a)} \widehat{\mathbf{E}}(\mathbf{r}_{a}).
$$
\n(2.4)

In this equation $\widehat{\mathbf{d}}^{(a)}$ is the dipole momentum operator of the atom *a*, $\mathbf{E}(\mathbf{r}_a)$ is the electric field operator, and \mathbf{r}_a is the position of the atom *a*.

The electric field operator $\mathbf{E}(\mathbf{r})$ in a microcavity can be obtained in a standard way by quantization of the classical field **E**(**r***,t*). The latter is a solution of Maxwell equations with corresponding boundary conditions.

Let us consider a coordinate system with the *z* axis perpendicular to the mirrors and with reference point $z = 0$ at one mirror (see Fig. 1). In such a case the boundary conditions can be written as follows: $E_x|_{z=0} = E_x|_{z=0}$ $E_y|_{z=0} = E_y|_{z=d} = 0$. We will use cgs units throughout the paper. Solving the Maxwell equations, we have

$$
\mathbf{E}(\mathbf{r},t) = \sum_{\mathbf{k},\alpha} \frac{i\omega_k}{c} b_{\mathbf{k},\alpha}(t) \mathbf{A}_{\mathbf{k},\alpha}(\mathbf{r}) + \text{c.c.},\tag{2.5}
$$

$$
b_{\mathbf{k},\alpha}(t) = b_{\mathbf{k},\alpha} \exp(-i\omega_k t),
$$
 (2.6)

$$
\mathbf{A}_{\mathbf{k},\alpha}(\mathbf{r}) = A_{\mathbf{k},\alpha}^0 \exp(i\mathbf{k}_{\shortparallel}\mathbf{r}_{\shortparallel}) \{ \mathbf{e}_x u_{\mathbf{k},\alpha}^x \sin(k_n z) \n+ \mathbf{e}_y u_{\mathbf{k},\alpha}^y \sin(k_n z) + \mathbf{e}_z u_{\mathbf{k},\alpha}^z \cos(k_n z) \}.
$$
 (2.7)

Here \mathbf{e}_x , \mathbf{e}_y , and \mathbf{e}_z are unit vectors of the chosen coordinate system; $\mathbf{r}_{\shortparallel} = x\mathbf{e}_x + y\mathbf{e}_y$, $\mathbf{k}_{\shortparallel} = k_x\mathbf{e}_x + k_y\mathbf{e}_y$, $k_n = \pi n/d$, $n =$ $0,1,2,\ldots$, and $\mathbf{u}_{\mathbf{k},\alpha} = \mathbf{e}_x u_{\mathbf{k},\alpha}^x + \mathbf{e}_y u_{\mathbf{k},\alpha}^y + \mathbf{e}_z u_{\mathbf{k},\alpha}^z$ is the unit polarization vector.

From the equation $\vec{\nabla} \cdot \mathbf{E}(\mathbf{r},t) = 0$ we obtain $k_x u_{\mathbf{k},\alpha}^x$ + $k_y u_{\mathbf{k},\alpha}^y + i k_n u_{\mathbf{k},\alpha}^z = 0$. The modified polarization vectors $\mathbf{u}_{\mathbf{k},\alpha}^{\prime} = \mathbf{e}_x u_{\mathbf{k},\alpha}^x + \mathbf{e}_y u_{\mathbf{k},\alpha}^y + \mathbf{e}_z i u_{\mathbf{k},\alpha}^z$ are orthogonal to the wave vector $\mathbf{k} = \mathbf{k}_{\text{H}} + \mathbf{e}_{z}k_{n}$ and obeys the following transferability condition:

$$
\sum_{\alpha} (u'_{\mathbf{k},\alpha})_{\mu} (u'_{\mathbf{k},\alpha})_{\nu}^{*} = \delta_{\mu\nu} - \frac{k_{\mu}k_{\nu}}{k^{2}}.
$$
 (2.8)

Here μ and ν denote the vector projection on the coordinate axes; the sum in (2.8) is over two orthogonal components of the modified polarization vectors.

To obtain the explicit expression of the Schrodinger electric field operator we make the standard replacement $b_{\mathbf{k},\alpha}(t) \rightarrow$ */*_{2*ωk*} $\hat{a}_{k,\alpha}$; $b_{k,\alpha}^*$ (*t*) → $\sqrt{\hbar/2\omega_k}\hat{a}_{k,\alpha}^{\dagger}$, where $\omega_k = ck$ is the photon frequency, and $\hat{a}_{\mathbf{k},\alpha}$ and $\hat{a}_{\mathbf{k},\alpha}^{\dagger}$ are the annihilation and creation operators. By this means the electric field operator inside a Fabry-Perot cavity can be presented as follows:

$$
\widehat{\mathbf{E}}(\mathbf{r}) = \sum_{\mathbf{k},\alpha} \frac{i\omega_k}{c} \sqrt{\frac{\hbar}{2\omega_k}} \widehat{a}_{\mathbf{k},\alpha} A_{\mathbf{k},\alpha}^0
$$
\n
$$
\times \left\{ \mathbf{e}_x u_{\mathbf{k},\alpha}^x \sin(k_n z) + \mathbf{e}_y u_{\mathbf{k},\alpha}^y \sin(k_n z) + \mathbf{e}_z u_{\mathbf{k},\alpha}^z \cos(k_n z) \right\} \exp(i\mathbf{k}_\parallel \mathbf{r}_\parallel) + \text{H.c.} \tag{2.9}
$$

Using the standard relation between the electric and magnetic field, we obtain the magnetic field operator,

$$
\widehat{\mathbf{H}}(\mathbf{r}) = \sum_{\mathbf{k},\alpha} \sqrt{\frac{\hbar}{2\omega_k}} \widehat{a}_{\mathbf{k},\alpha} A_{\mathbf{k},\alpha}^0 \left\{ \left(i k_y u_{\mathbf{k},\alpha}^z - k_n u_{\mathbf{k},\alpha}^y \right) \cos(k_n z) \mathbf{e}_x \right. \n+ \left(k_n u_{\mathbf{k},\alpha}^x - i k_x u_{\mathbf{k},\alpha}^z \right) \cos(k_n z) \mathbf{e}_y \n+ \left(i k_x u_{\mathbf{k},\alpha}^y - i k_y u_{\mathbf{k},\alpha}^x \right) \sin(k_n z) \left\} \exp(i \mathbf{k}_{\shortparallel} \mathbf{r}_{\shortparallel}) + \text{H.c.}
$$
\n(2.10)

Note that in this paper we use cgs units and so we apply the notation **H** for the microscopic magnetic field in accordance with the standard text book $[49]$. In (2.9) and (2.10) $A_{\mathbf{k},\alpha}^{0}$ is the normalization constant. It can be calculated on the basis of the standard form of the field Hamiltonian,

$$
\widehat{H}_{f} = \int_{V_{q}} \frac{1}{8\pi} (\widehat{\mathbf{E}}^{2} + \widehat{\mathbf{H}}^{2}) dV
$$

$$
= \sum_{\mathbf{k}, \alpha} \hbar \omega_{k} \left(\widehat{a}^{\dagger}_{\mathbf{k}, \alpha} \widehat{a}_{\mathbf{k}, \alpha} + \frac{1}{2} \right). \tag{2.11}
$$

Here V_q is the quantization volume, $V_q = \{0 \leq z \leq d\} \times \{0 \leq z \leq d\}$ $x, y \leq L$. From Eq. (2.11) we have

$$
A_{\mathbf{k},\alpha}^{0} = \sqrt{\frac{8\pi c^2}{L^2 d}} \times \begin{cases} 1, & \text{if } n \in N \\ 1/\sqrt{2}, & \text{if } n = 0. \end{cases}
$$
 (2.12)

L is the longitudinal size of the quantization volume.

In accordance with $[41]$ and $[42]$ we will seek the wave function ψ as an expansion in a set of eigenfunctions ψ_l of the operator H_0 :

$$
\psi = \sum_{l} b_l(t)\psi_l. \tag{2.13}
$$

Here, the subscript *l* defines the state of all atoms and the field. Using this representation of the wave function we convert the equation [\(2.1\)](#page-1-0) to the system of linear differential equations for the quantum amplitudes,

$$
i\hbar \frac{\partial b_l(t)}{\partial t} - E_l b_l(t) = \sum_j V_{lj} b_j(t). \tag{2.14}
$$

In this equation E_l is the energy of the l state of the system, which consists of noninteracting atoms and electromagnetic field.

Because of the infinity number of the field states the total number of equations in the system (2.14) is equal to infinity.

The key simplification of the approach employed is in the restriction of the total number of states $|l\rangle$ taken into account.

We will calculate all radiative correction up to the second order of the fine structure constant. In this case we can consider only the following states (see $[50]$):

(1) Onefold atomic excited states,

$$
\psi_{e_a} = |g,...,g,e,g,...,g\rangle \otimes |vac\rangle, E_{e_a} = \hbar\omega_a.
$$

(2) Resonant single-photon states,

$$
\psi_g = |g,...,g\rangle \otimes |\mathbf{k},\alpha\rangle, E_g = \hbar\omega_k.
$$

(3) Nonresonant states with two excited atoms and one photon,

 $\psi_{e_a e_b} = |g,...,g,e,g,...,g,e,g,...,g\rangle \otimes |\mathbf{k},\alpha\rangle, E_{e_a e_b} = \hbar(\omega_a +$ ω_b) + $\hbar \omega_k$.

In the rotating wave approximation it is enough to take into account only the first and second group of states. Nonresonant states are necessary for a correct description of the dipoledipole interaction at short interatomic distances, comparable with λ_0 .

For a description of the coherent external light scattering, it is necessary to complete the set of quantum states by the vacuum state without excitation both in the atomic and field subsystem,

 $\psi_{g'} = |g,...,g\rangle \otimes |vac\rangle, \quad E_{g'} = 0.$

In the framework of the assumptions considered here, the quantum amplitude of the state ψ_{g} does not change during the evolution of the system. It is explained by the fact that any transitions between ψ_{g} and the other quantum states taken into account are impossible. The Lamb shift is considered to be included in ω_0 .

Despite the restriction of the total number of quantum states, the set of equations remains infinite.We can, however, formally solve it without any additional approximations. For this we express the amplitudes of quantum states with one photon via the amplitudes of atomic excitation. Then we put it in the equations for amplitudes of atomic excitation. Thus, we obtain a closed finite system of equations for onefold excited states of atomic subsystem b_e . For Fourier components $b_e(\omega)$ we have (at greater length see [\[42\]](#page-9-0))

$$
\sum_{e'} [(\omega - \omega_a) \delta_{ee'} - \Sigma_{ee'}(\omega)] b_{e'}(\omega) = i \delta_{e'o}.
$$
 (2.15)

This specific set of equations was obtained under the assumption that at the initial time only one atom is excited. We denote it by the subscript *o*. All other atoms are in the ground states at $t = 0$ and electromagnetic field is in the vacuum state. The system (2.15) with the initial conditions considered here allows us to analyze both stationary light scattering as well as nonstationary problems (see [\[42\]](#page-9-0)).

The size of the system (2.15) is determined by the number of atoms *N* and the structure of their energy levels. In this paper we consider the impurity centers with ground state $J = 0$. Total angular momentum of the excited state is $J = 1$. It includes three sublevels $e = |J,m\rangle$, which differ by the value of angular momentum projection on the quantization axis $m = -1, 0, 1$. Therefore, the total number of onefold atomic excited states is 3*N*. This scheme of levels corresponds to atoms with two valence electrons such as Sr, Yb, Ca.

The matrix $\Sigma_{ee}(\omega)$ describes both spontaneous decay and excitation exchange between the atoms. This matrix can be calculated as follows:

$$
\Sigma_{ee'}(\omega) = \sum_{g} V_{e;g} V_{g;e'} \zeta(\hbar \omega - E_g)
$$

$$
+ \sum_{ee} V_{e;ee} V_{ee;e'} \zeta(\hbar \omega - E_{ee}). \qquad (2.16)
$$

In this equation $\zeta(x)$ is a singular function which is determined by the relation $\zeta(x) = \lim_{k \to \infty} (1 - \exp(ikx))/x$.

We will calculate the sum over the field variables in Eq. (2.16) in the limit $L \to \infty$. This implies summation over *n* (k_n) , the integration over k_{\parallel} , and the polar angle φ as well as summation over polarization types in accordance with following:

$$
\sum_{g} \text{or} \sum_{ee} \rightarrow \frac{L^2}{(2\pi)^2} \sum_{n=0}^{+\infty} \left(\int_0^{+\infty} k_{\shortparallel} dk_{\shortparallel} \int_0^{2\pi} d\varphi \sum_{\alpha} .
$$

The prime sign here denotes an additional coefficient 1*/*2 in the sum over *n* for $n = 0$. This coefficient appears from Eq. [\(2.12\)](#page-2-0).

When calculating matrix elements of the operator V in (2.16) we will denote by index *a* those atoms which transit from excited state to ground one and by index *b* atoms which perform reverse transition. With Eqs. (2.9) and (2.12) we have

$$
V_{e,g} = \langle e | \widehat{V} | g \rangle = -\mathbf{d}_{e_b;g_b} i \sqrt{\frac{4\pi \hbar \omega_k}{L^2 d}}
$$

$$
\times \{ \mathbf{e}_x u_{\mathbf{k},\alpha}^x \sin(k_n z_b) + \mathbf{e}_y u_{\mathbf{k},\alpha}^y \sin(k_n z_b) + \mathbf{e}_z u_{\mathbf{k},\alpha}^z \cos(k_n z_b) \} \exp(i \mathbf{k}_{\shortparallel} \mathbf{r}_{\shortparallel b}), \qquad (2.17)
$$

$$
V_{g;e'} = \langle g | \widehat{V} | e' \rangle = \mathbf{d}_{g_a;e_a} i \sqrt{\frac{4\pi \hbar \omega_k}{L^2 d}}
$$

$$
\times \{ \mathbf{e}_x (u_{\mathbf{k},\alpha}^x)^* \sin(k_n z_a) + \mathbf{e}_y (u_{\mathbf{k},\alpha}^y)^* \sin(k_n z_a)
$$

$$
+ \mathbf{e}_z (u_{\mathbf{k},\alpha}^z)^* \cos(k_n z_a) \} \exp(-i \mathbf{k}_{\shortparallel} \mathbf{r}_{\shortparallel a}). \qquad (2.18)
$$

The calculation of $V_{e;ee}$ $V_{ee;e'}$, which can be performed in the same way, gives $V_{e;ee} = V_{e;g}$, $V_{ee;e'} = V_{g;e'}$.

For arbitrary *ω*, the explicit expression of the matrix $\Sigma_{ee}(\omega)$ is very complicated. We can, however, simplify it essentially under so-called pole approximation when its value for frequency ω is replaced by its value for frequency ω_0 of the atomic resonance. This approximation was studied in detail in [\[51\]](#page-9-0), where it was shown that it can be applied in systems where retardation effects are insignificant. This condition can be satisfied in the real experiment with a good accuracy even for dense atomic ensembles. In the pole approximation we get

$$
\Sigma_{ee'}(\omega_0) = \frac{L^2}{4\pi^2} \sum_{n=0}^{+\infty} \int_0^{+\infty} k_n dk_n \int_0^{2\pi} d\varphi \frac{4\pi \hbar \omega_k}{L^2 d} \left\{ d_{e_b,g_b}^x d_{g_a;e_a}^x \sin(k_n z_b) \sin(k_n z_a) \left(\frac{k_y^2 + k_n^2}{k^2} \right) \right.\n+ d_{e_b;g_b}^x d_{g_a;e_a}^y \sin(k_n z_b) \sin(k_n z_a) \left(-\frac{k_x k_y}{k^2} \right) + d_{e_b;g_b}^x d_{g_a;e_a}^z \sin(k_n z_b) \cos(k_n z_a) \left(-i \frac{k_x k_n}{k^2} \right) \right.\n+ d_{e_b;g_b}^y d_{g_a;e_a}^x \sin(k_n z_b) \sin(k_n z_a) \left(-\frac{k_x k_y}{k^2} \right) + d_{e_b;g_b}^y d_{g_a;e_a}^y \sin(k_n z_b) \sin(k_n z_a) \left(\frac{k_x^2 + k_n^2}{k^2} \right) \right.\n+ d_{e_b;g_b}^y d_{g_a;e_a}^z \sin(k_n z_b) \cos(k_n z_a) \left(-i \frac{k_y k_n}{k^2} \right) + d_{e_b;g_b}^z d_{g_a;e_a}^x \cos(k_n z_b) \sin(k_n z_a) \left(i \frac{k_x k_n}{k^2} \right) \right.\n+ d_{e_b;g_b}^z d_{g_a;e_a}^y \cos(k_n z_b) \sin(k_n z_a) \left(i \frac{k_y k_n}{k^2} \right) + d_{e_b;g_b}^z d_{g_a;e_a}^z \cos(k_n z_b) \cos(k_n z_a) \left(\frac{k_x^2 + k_y^2}{k^2} \right) \right\} \times \exp(i \mathbf{k}_{\text{ir}} \mathbf{r}_{\text{lab}}) \left[-i \pi \delta(\hbar \omega_0 - \hbar \omega_k) - i \pi \delta(-\hbar \omega_0 - \hbar \omega_k) + \text{v.p.} \left(\frac{1}{\hbar \omega_0 - \hbar \omega_k} + \frac{1}{-\hbar \omega_0 - \hbar \omega_k} \right) \right].
$$
 (2.19)

In this equation $\mathbf{r}_{\text{sub}} = \mathbf{r}_{\text{sub}} - \mathbf{r}_{\text{max}}$, the sum over polarization types was calculated using the relation [\(2.8\)](#page-1-0). The singular ζ function is represented as follows $\zeta(x) = -i\pi\delta(x) + \text{v.p.}/x$, where v.p. means the principal value of the integral which contains the *ζ* function.

The diagonal element of the matrix (2.19) describes the Lamb shift and the natural linewidth of an atom inside a cavity. The dipole approximation used here does not allow us to calculate the Lamb shift correctly. This manifests itself in the infinity real part of the diagonal element. We can, however, consider that the Lamb shift is included into *ω*0. Hereafter we will associate *ω*⁰ with the resonant transition frequency taking into account the Lamb shift.

The imaginary part of the diagonal element determining the natural linewidth can be calculated as follows:

$$
\Sigma_{ee'}(\omega_0)\Big|_{e=e'} = -\frac{i\pi}{d} d_{ea;ga}^z d_{ga;ea}^z \frac{\omega_0^2}{c^2} - \frac{i\pi}{d} \sum_{n=1}^{[\frac{\omega_0 d}{\pi c}]} \left\{ \left(\frac{\omega_0^2}{c^2} + k_n^2\right) \sin^2(k_n z_a) \left(d_{ea;ga}^x d_{ga;ea}^x + d_{ea;ga}^y d_{ga;ea}^y\right) + 2\left(\frac{\omega_0^2}{c^2} - k_n^2\right) \cos^2(k_n z_a) d_{ea;ga}^z d_{ga;ea}^z \right\}.
$$
\n(2.20)

Double brackets here means the integer part. The natural linewidth *γ* can be obtained using the relation $\Sigma_{ee}(\omega_0)|_{e=e'}=$ −*iγ/*2; see [\[42\]](#page-9-0) for detail.

If *e'* and *e* correspond to excited states of different atoms, for example, atoms *a* and *b*, matrix element $\Sigma_{ee}(\omega)$ describes excitation exchange between these atoms. As it is known this exchange is responsible for interatomic dipole-dipole interaction. The matrix element $\Sigma_{ee}(\omega)$ is easy to calculate in the coordinate frame with the *X* axis along the vector **r**_{nab}. In this frame $k_x = k_0 \cos \varphi$, $k_y = k_0 \sin \varphi$, and $\mathbf{k}_0 \mathbf{r}_{0ab} =$ $k_{\parallel}r_{\parallel ab}\cos\varphi$. The double integral in Eq. [\(2.19\)](#page-3-0) can be simplified

by the following relations:

$$
k_x \exp(i\mathbf{k}_{||}\mathbf{r}_{||ab}) = -i\frac{\partial}{\partial x_{ab}} \exp(i\mathbf{k}_{||}\mathbf{r}_{||ab}),
$$

\n
$$
k_x^2 \exp(i\mathbf{k}_{||}\mathbf{r}_{||ab}) = -\frac{\partial^2}{\partial x_{ab}^2} \exp(i\mathbf{k}_{||}\mathbf{r}_{||ab}),
$$

\n
$$
k_x k_y \exp(i\mathbf{k}_{||}\mathbf{r}_{||ab}) = -\frac{\partial^2}{\partial x_{ab} y_{ab}} \exp(i\mathbf{k}_{||}\mathbf{r}_{||ab}).
$$

Here $x_{ab} = x_b - x_a$, $y_{ab} = y_b - y_a$. For the other items in [\(2.19\)](#page-3-0) we have similar relations, and we have

$$
\Sigma_{ee'}(\omega_0)\Big|_{a\neq b} = \sum_{n=0}^{+\infty} \left(\widehat{A}_n \int_0^{+\infty} k_{\shortparallel} dk_{\shortparallel} \int_0^{2\pi} d\varphi \frac{c}{\pi d} \frac{1}{k} \times \exp(ik_{\shortparallel} r_{\shortparallel ab} \cos \varphi) \Big[-i\pi \delta(\omega_0 - ck) + \frac{2ck}{\omega_0^2 - c^2 k^2} \Big].
$$
\n(2.21)

The differential operator A_n is determined as follows:

$$
\widehat{A}_{n} = d_{e_{b};g_{b}}^{x} d_{g_{a};e_{a}}^{y} \sin(k_{n}z_{b}) \sin(k_{n}z_{a}) \left(\frac{\partial^{2}}{\partial x \partial y}\right) + d_{e_{b};g_{b}}^{x} d_{g_{a};e_{a}}^{x} \sin(k_{n}z_{b}) \sin(k_{n}z_{a}) \left(k_{n}^{2} - \frac{\partial^{2}}{\partial y^{2}}\right) \n+ d_{e_{b};g_{b}}^{x} d_{g_{a};e_{a}}^{z} \sin(k_{n}z_{b}) \cos(k_{n}z_{a}) \left(-k_{n} \frac{\partial}{\partial x}\right) + d_{e_{b};g_{b}}^{y} d_{g_{a};e_{a}}^{x} \sin(k_{n}z_{b}) \sin(k_{n}z_{a}) \left(\frac{\partial^{2}}{\partial x \partial y}\right) \n+ d_{e_{b};g_{b}}^{y} d_{g_{a};e_{a}}^{y} \sin(k_{n}z_{b}) \sin(k_{n}z_{a}) \left(-\frac{\partial^{2}}{\partial x^{2}} + k_{n}^{2}\right) + d_{e_{b};g_{b}}^{y} d_{g_{a};e_{a}}^{z} \sin(k_{n}z_{b}) \cos(k_{n}z_{a}) \left(-k_{n} \frac{\partial}{\partial y}\right) \n+ d_{e_{b};g_{b}}^{z} d_{g_{a};e_{a}}^{x} \cos(k_{n}z_{b}) \sin(k_{n}z_{a}) \left(k_{n} \frac{\partial}{\partial x}\right) - d_{e_{b};g_{b}}^{z} d_{g_{a};e_{a}}^{z} \cos(k_{n}z_{b}) \cos(k_{n}z_{a}) \left(\frac{\partial^{2}}{\partial x^{2}} + \frac{\partial^{2}}{\partial y^{2}}\right) \n+ d_{e_{b};g_{b}}^{z} d_{g_{a};e_{a}}^{y} \cos(k_{n}z_{b}) \sin(k_{n}z_{a}) \left(k_{n} \frac{\partial}{\partial y}\right).
$$
\n(2.22)

Here $x = x_b - x_a$, $y = y_b - y_a$.

The calculation of the double integral in Eq. (2.21) produces Bessel functions J_0 , K_0 , and N_0 . Thus, we have

$$
\Sigma_{ee'}(\omega_0)\Big|_{a\neq b} = \frac{2\pi}{d} \sum_{n=0}^{\lfloor \frac{\omega_0 d}{2C} \rfloor} \hat{A}_n
$$
\n
$$
\times \left[N_0 \left(r_{\text{sub}} \sqrt{\frac{\omega_0^2}{c^2} - k_n^2} \right) - i J_0 \left(r_{\text{sub}} \sqrt{\frac{\omega_0^2}{c^2} - k_n^2} \right) \right] - \frac{4}{d} \sum_{n=\lfloor \frac{\omega_0 d}{2C} \rfloor + 1}^{+\infty} \hat{A}_n K_0 \left(r_{\text{sub}} \sqrt{k_n^2 - \frac{\omega_0^2}{c^2}} \right). \tag{2.23}
$$

 $\sqrt{ }$

The differential operator A_n yields bulky expressions which we do not show here.

The explicit expressions (2.20) and (2.23) for the matrix $\Sigma_{ee}(\omega_0)$ obtained in this section allows us to solve the set of equations [\(2.15\)](#page-2-0) numerically and obtain, on this background, the Fourier amplitudes of atomic states $b_e(\omega)$. Using $b_e(\omega)$ we can obtain the amplitudes of all states taken into account in our calculations (see $[42]$) and, consequently, the wave function of the considered system. Note that expressions [\(2.20\)](#page-3-0) and (2.23) describing spontaneous decay and excitation exchange between impurity centers inside the cavity essentially differ from those for atomic ensemble in free space. These differences lead to some peculiarities of collective polyatomic effects in dense atomic systems located into cavity.

In the next section, we will use the obtained general equations to calculate the transition spectrum of an excited atom and the time dependence of the total excitation of atomic ensemble. On this basis, we will analyze radiation trapping in the considered system.

3. RESULTS AND DISCUSSION

The influence of the dipole-dipole interaction on the properties of atomic ensemble is determined not only by the atomic density. The shifts of the energy levels caused by the internal fields of a dielectric are also very important. The value of these shifts depends on a number of factors, first of all, on the nature of the dielectric and its temperature. As it was

mentioned above in the present paper we will assume that the temperature is low enough to neglect the electron-phonon interaction. So the spectral lines of impurity centers are zero phonon.

The shift of the transition line can be presented as a sum of its average value Δ and some random contribution connected with the inhomogeneity of the internal fields of the dielectric.We consider this random contribution to be normally distributed with rms deviation *δ*. The ratio of *δ* to natural line width of the atoms γ_0 characterizes the degree of resonance between impurity atoms. This is one of the key parameters of the considered system in our theory.

Depending on the symmetry of the internal fields of a dielectric, the average shift $\overline{\Delta}$ can be both the same for all Zeeman sublevels of the excited state and different. In general, the theory allows us to analyze both cases. Only for specifics, in this section we limit ourselves to the first case. This corresponds to the cubic symmetry of internal fields of a medium, for instance. Hereafter we will consider Δ to be included in the resonant transition frequency ω_0 .

In the framework of the general approach we can consider an arbitrary distance between the mirrors of a Fabry-Perot cavity. However, the most exciting case is $d < \lambda_0/2$ due to the suppression of the spontaneous decay of the states $m = \pm 1$ in the cavity. So we focus our attention on this case. Taking k_0^{-1} as a unit of length, hereafter we consider $d = 3$.

In this paper we assume spatially localized initial excitation of the ensemble. Such initial condition can be prepared by a two-photon resonance method. In the framework of this method the sample is illuminated by two narrow and off-resonant orthogonally propagated light beams (both beams parallel to the mirrors of a cavity). Each beam does not cause single-photon excitation, but their simultaneous interaction with atoms in the crossing region cause two-photon excitation from the ground *S* to the high-energy excited *D* state if conditions of two-photon resonance are satisfied. If the transition frequency from the high-energy *D* state to the studied *P* state is high enough so that its resonant wavelength $\lambda_{D\rightarrow P}$ < 2*d*, this spontaneous transition leads to population of the *P* state. Note that the cascade transition from *D* to *S* state can be forbidden, in particular, due to the spontaneous decay suppression in each step of the cascade. So the registration of the photon resonant to the transition $D \longrightarrow P$ means the *P* state population.

The thereby described method allows obtaining a small cluster of excited atoms in the middle of the sample. For simplicity thereafter in the paper we will consider that at the initial time only one atom located in the center of a sample is excited. Note that the possibilities of two-photon excitation $5s \text{ } S-2(1/2) \longrightarrow 5p \text{ } P-2(j) \longrightarrow 5d \text{ } D-2(j)' \text{ of } \text{rubidium}$ atoms have been already studied in Ref. [\[52\]](#page-9-0).

Equation (2.20) shows that the natural linewidth of the excited atom inside the cavity depends on its *z* position even in the case of a single atom. So all the results must depend on this parameter. In the framework of the general theory we can consider an arbitrary position of all the atoms, including the excited initially atom. From the experimental point of view the position of the excited atom is determined by the crossing region of two beams. Further we will consider $z_{\rm exc} = d/2.$

Note that the matrix $\Sigma_{ee'}$ and subsequently any physical observable depends on the positions of all impurity atoms. In this paper we analyze spatially disordered atomic ensembles with uniform (on average) distribution of atomic density. So we average all the results over random spatial configurations of the ensemble as well as over random shifts of energy levels caused by the inhomogeneity of the internal fields of a dielectric. This averaging is performed by a Monte Carlo method.

A. Atomic transition spectrum

Figure 2 shows the transition spectrum of the central atom, which is initially excited. The calculations were made for $\delta = 0$. In this case all the atoms are resonant to each other, so the role of the dipole-dipole interaction is maximal. Ensembles with two densities $n = 0.01$ and $n = 0.05$ are considered.

Density $n = 0.01$ is small and in this case cooperative effects in the free space manifest themselves slightly; see [\[53\]](#page-9-0) for detail. Difference between transition spectrum of the free atom and atom excited in the ensemble when the cavity is absent is very small. However, in a cavity the dipole-dipole

FIG. 2. Transition spectrum of an atom inside a microcavity. $d = 3$, $\delta = 0$. (a) Real part; (b) imaginary part. 1, $n = 0.01$, $m = \pm 1$; 2, $n = 0.01, m = 0; 3, n = 0.05, m = \pm 1; 4, n = 0.05, m = 0.$

interaction transforms the transition spectrum significantly. First of all, in Fig. [2](#page-5-0) we see that the transition spectrum in a cavity for Zeeman sublevels $m = \pm 1$ extremely differs from one for $m = 0$. Despite the strong cavity suppression of the spontaneous decay from sublevels $m = \pm 1$ [\[22\]](#page-9-0), we observe that in the ensemble of the density $n = 0.01$ the width of the transition spectrum is approximately equal to 0*.*6*γ*0. This broadening is determined completely by the polyatomic cooperative effect. Also we observe some blue shift as well as an essential discrepancy between the spectrum shape and a typical Lorentz profile. For $m = 0$ the modification of the spectrum shape is considerably less than for $m = \pm 1$. It can be explained by the fact that for $m = 0$ the spontaneous decay of a single atom in a cavity is not suppressed unlike sublevels $m = \pm 1$. However, a noticeable red shift is observed even in this case.

In the case of essentially higher density $n = 0.05$ the dipole-dipole interaction plays an important role for the atomic ensembles in free space, without cavity [\[8\]](#page-9-0). Nevertheless, Fig. [1](#page-1-0) shows that the microcavity modifies the transition spectrum additionally. In Fig. [2](#page-5-0) one can see a blue shift comparable with γ_0 for $m = \pm 1$ and approximately the same red one for $m = 0$. The shape of the transition spectrum significantly differs from a Lorentz profile for any Zeeman sublevel due to an essential role of the dipole-dipole interaction. This interaction causes also broadening of the spectrum as density increases.

B. Time dependence of the total excitation of atomic system

The inverse Fourier transform of $b_e(\omega)$ allows us to obtain the time dependence of the quantum amplitudes of the onefold atomic excited states,

$$
b_e(t) = \int_{-\infty}^{+\infty} \frac{i d\omega}{2\pi} \exp(-i\omega t) R_{eo}(\omega).
$$
 (3.1)

Here matrix $R_{eo}(\omega)$ is the resolvent of the considered system projected on the onefold atomic excited states [\[42\]](#page-9-0). It is determined from Eq. [\(2.15\)](#page-2-0) as follows:

$$
R_{ee'}(\omega) = [(\omega - \omega_a)\delta_{ee'} - \Sigma_{ee'}(\omega_0)]^{-1}.
$$
 (3.2)

The total excited state population $P_{sum}(t)$ is given by a sum of $|b_e(t)|^2$ over all atoms in the ensemble. Besides $P_{\text{sum}}(t)$ we calculate the time-dependent collective decay rate:

$$
\gamma(t) = -\frac{1}{P_{\text{sum}}(t)} \frac{dP_{\text{sum}}(t)}{dt}.
$$
\n(3.3)

Figure 3 shows the time dependence of the total excited state population and the collective decay rate in the case $\delta = 0$ both for the ensemble in free space and in the cavity. The results are presented for the atomic density $n = 0.1$ and for the size a sample $R = 14$. *R* means the radius of the spherical sample in the case of free space and the radius of a cylindrical sample in the case of a cavity. First of all, we observe that the total excited state population in the case of a microcavity decreases slower than one in the case of free space. Besides that, the decay rate of sublevels $m = \pm 1$ in a cavity is less than of the Zeeman sublevel $m = 0$. It is connected with mentioned features of the field modes structure in the microcavity.

FIG. 3. Time dependence of the total excited state population (a) and collective decay rate (b). $n = 0.1$, $\delta = 0$, $R = 14$. 1, free space; 2, microcavity, $d = 3$, $m = \pm 1$; 3, microcavity, $d = 3$, $m = 0$.

For the time interval $t \gg \gamma_0^{-1}$ the time dependence of the total excited state population in the semilogarithmic scale is close to linear, and subsequently the collective decay rate depends on time weakly. This case is similar to a Holstein mode decay. For $t \sim \gamma_0^{-1}$ the time dependence of the total excited state population is more complex because both superradiant and subradiant collective states influence it. For the atomic ensemble in free space we observe that the collective decay rate decreases with time here. It can be explained by the fact that the influence of superradiant states decreases with time whereas the influence of subradiant states increases [\[8\]](#page-9-0). The same holds true for the microcavity in the case of sublevel $m = 0$ decay. However, the time dependence of the collective decay rate of Zeeman sublevels $m = \pm 1$ in the microcavity is not a monotonic function, and it has a maximum at $t = 1.2\gamma_0^{-1}$ for considered parameters. To understand this effect we studied the spectral distribution of the density of collective states. The performed analysis shows that the frequency distribution of the density of states with proper lifetimes has two peaks. It causes the quantum beats which manifest themselves in the excited state population. Generally,

FIG. 4. The time of radiation trapping in the microcavity. $n = 0.1$, $d = 3, m = \pm 1.$

quantum beats influence the collective decay rate both for $m = \pm 1$ and $m = 0$. However, for $m = \pm 1$ the role of the described mechanism is more significant, which is connected with the suppression of the spontaneous decay of a single atom in the microcavity.

C. The time of radiation trapping

We will estimate the typical time of radiation trapping *τ* from the relation $P_{sum}(\tau) = 1/e$. In this section we will concentrate our attention on the case of Zeeman sublevels $m = \pm 1$ initial excitation in view of the fact that it provides radiation trapping longer than that corresponding to $m = 0$. In addition, we point out that the time of radiation trapping in the microcavity is usually bigger than in the case of the atomic ensemble with the same density in free space. For example, in the case $n = 0.1$, $R = 14$, $\delta = 0$ we have $\tau = 60\tau_0$ in a cavity $(m = \pm 1)$, whereas $\tau = 25\tau_0$ in a free space. Here $\tau_0 = 1/\gamma_0$ is the natural lifetime of excited states of the free atom.

Figure $4(a)$ shows τ depending on the size of a sample. The atomic density is chosen $n = 0.1$. The results are presented for different values of the rms deviation of the inhomogeneous shifts of the resonant transition frequency. The dependence $\tau(R)$ is complex but as the size of the system increases it approaches to parabola. In the case of mutually resonance impurities $\delta = 0$ we observe dependence close to quadratic $\tau/\tau_0 \propto R^2$ starting approximately with $R \approx 15$. It corresponds to the case when the size of the system is much greater than photon mean free path *l*ph. The latter can be estimated on the basis of calculation [\[43–44\]](#page-9-0) as *l*ph ∼ 1*.*6. Observed quadratic dependence is typical for diffuse radiation transfer.

As δ increases, the mean free path of the photon also increases. In the case $\delta \neq 0$ the regime close to diffuse radiation transfer is achieved for bigger systems. The dependence $\tau(\delta)$ for $R = 14$ is shown in Fig. 4(b). The character of this dependence is determined by two different factors. On the one hand, radiation trapping is connected with the cooperative multiple scattering. The influence of this mechanism decreases with increasing in *δ*. On the other hand, trapping time depends on the spontaneous decay suppression, which manifests itself more noticeably as *δ* increases. For small *δ* the first mechanism is more significant, so *τ* (*δ*) decreases. In the case of large *δ* decay suppression dominates; this leads to increasing in *τ* . Generally, the dependence $\tau(\delta)$ has a minimum.

Nonresonant impurity centers (δ **-** *γ***0***)*

In a range of solid dielectrics the shifts of resonant transition frequency of impurity centers *δ* are relatively large. For instance, it is typical for NV centers in a diamond. If $\delta \gg \gamma_0$ the average cross section associated with an individual atom is much less than λ_0^2 . In such a case the dipole-dipole interaction can be significant only for high density of impurities, when the average distance between mutually resonant atoms $(n\gamma_0/\delta)^{-1/3}$ is less or comparable with wavelength λ_0 or to put it differently when mean free path of photon $l_{ph} = (n\sigma_0\gamma_0/\delta)^{-1}$ satisfies the inequality $l_{\text{ph}} \le \lambda_0$. Here $\sigma_0 = 3\lambda_0^2/2\pi$ is the resonant cross section concerning the free atom.

Assuming the random inhomogeneous shifts of the transition frequency to be normally distributed, we have

$$
\frac{dn}{d\Delta} = \frac{n_f}{\delta\sqrt{2\pi}} \exp\left(-\frac{\Delta^2}{2\delta^2}\right).
$$
 (3.4)

In this equation n_f means the total density of impurity atoms.

In the considered case $\delta \gg \gamma_0$ not all the atoms in the ensemble essentially influence on the radiative processes but only those which have the inhomogeneous shifts close to that of the initially excited atom. We will denote the latter as Δ_e . In our calculation we take into consideration only atoms with inhomogeneous shifts $\Delta \in [\Delta_e - \Delta_1; \Delta_e + \Delta_1]$, where Δ_1 is some computational cutoff frequency. We choose Δ_1 so big that the obtained results do not change with further increasing of this parameter.

Figure [5](#page-8-0) shows the dependence of the time of radiation trapping on the size of a sample for two different densities $n_f = 5$ and $n_f = 2$. The rms deviation of the inhomogeneous shifts is $\delta = 10^3 \gamma_0$. The mean free path of a photon can be estimated as $l_{ph} = 10.6$ and $l_{ph} = 26.5$ for $n_f = 5$ and $n_f = 2$, respectively. In these cases the investigation of the dependence $\tau(R)$ in the regime of diffuse radiation transfer $R \gg l_{\text{ph}}$ is connected with essential computational difficulties. We limit ourselves by the case $R \ge l_{ph}$.

FIG. 5. The time of radiation trapping in the microcavity in the case of nonresonant impurity centers. $\delta = 10^3 \gamma_0$, $d = 3$, $m = \pm 1$.

Figure 5 demonstrates very weak dependence of the time of radiation trapping on the size of the sample. It is explained by competition of two effects. The first one is spontaneous decay suppression and the second is trapping under multiple scattering. The role of multiple scattering increases with size, whereas the spontaneous decay is substantially suppressed for small *R*. This suppression explains also increasing of trapping time with density decreasing. As we approach the single atom limit the time of radiation trapping increases up to infinity.

4. CONCLUSION

We have developed a consistent quantum mechanical theory of cooperative effects in ensembles of pointlike impurity centers imbedded into transparent dielectric and located in a Fabry-Perot cavity with perfectly conducting plates. Our approach is based on the solution of the non-steady-state Schrodinger equation for the wave function of the joint system consisting of an ensemble of motionless atoms and a weak electromagnetic field. The interaction of impurity atoms with the dielectric is simulated by introduction inhomogeneous shifts of the atomic energy levels. The general approach allows us to analyze atomic ensembles with arbitrary shape and spatial distribution of impurities.

As an example of a practical implementation of this approach in the present work we study the simultaneous influence of the cavity and resonant dipole-dipole interaction on the shape of the line of atomic transition as well as on light trapping in dense impurity ensembles. We analyze this influence depending on the size of the ensembles, its density, as well as on the rms deviation of the transition frequency shifts caused by the symmetry disturbance of the internal fields of the dielectric medium. The special attention is given to the case when the distance between two mirrors is less than a half of the transition wavelength. This case is of particular interest due to practically complete suppression of spontaneous decay of some Zeeman sublevels of the atomic exited state.

In our opinion, the theory described in the present paper can be further used for the investigation of Anderson localization of light in the ensembles of impurity centers. It can be done on the basis of the spectral analysis of the collective states in such ensembles [\[54,55\]](#page-9-0). A special attention here should be paid to the case when the average shift of the transition frequency of impurity atoms caused by the internal fields of a dielectric $\overline{\Delta}$ is different for different Zeeman sublevels. In this case the excited state is not degenerate, which promotes the Anderson localization [\[55\]](#page-9-0).

The developed theory can be generalized to the case of the real susceptibility of the dielectric. In addition, it can be further generalized to the atomic ensembles in the waveguide. The case when the resonant frequency of atomic transition is less than the cutoff frequency of the waveguide attracts particular interest due to spontaneous decay suppression of all the Zeeman sublevels. Moreover, the analysis of the atomic systems in a waveguide can be useful for the investigation of Anderson localization, because in quasi-1D systems all the collective states are localized [\[56,57\]](#page-9-0).

In the present work we restrict our consideration by the case of a very weak electromagnetic field. We assume that the field is so weak that we can take into account the states with no more than one photon. Under this approximation we neglect all the nonlinear effects. One more way to generalize the described theory is to consider more than one photon in the field subsystem. It will allow us to analyze the influence of the dipole-dipole interaction on the statistics of light as well as the atomic excitation blockade.

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