Peculiarities of excitation trapping in dense polyatomic ensemble in a Fabry-Perot cavity

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Abstract. We report the investigation of the peculiarities of excitation trapping caused by the cooperative effects in a disordered ensemble of point-like impurity centers (atoms) embedded in a transparent dielectric and located in a Fabry-Perot cavity. On the basis of the general quantum microscopic theory we have calculated cooperative spontaneous decay of an excited atom located in the center of a sample. It is shown that in the case when the distance between the mirrors of a cavity $d$ is less or comparable with resonant wavelength of the atomic transition $\lambda_0$, the character of cooperative effects in a cavity essentially differs from one without a cavity. For the opposite case $d \gg \lambda_0$ cooperative effects in a cavity manifest themselves almost like in the case of absence of a cavity.

1. Introduction

Since the seminal work of Purcell [1] light interface with atoms localized inside a cavity or waveguide as well as near its surface has attracted a considerable attention. Now it is understood that using of a cavity offers an exciting tool to control over the atomic radiative properties, in particular, the enhancement and inhibition of the spontaneous decay rate [2] – [5]. Furthermore, the spontaneous decay can be suppressed almost completely by choosing the appropriate geometry of a cavity. Light matter interaction in the presence of nanophotonic structures, such as nanofibers [6] – [8], photonic crystal cavities [9] and waveguides [10] – [11] propose future applications for quantum metrology, scalable quantum networks and quantum information science [12] – [13].

Besides the cavity, the interaction of active impurity atoms with the surrounding dielectric medium can influence on its radiative properties (see [14] – [24] and references therein). Even in the case of a transparent dielectric the internal fields of a medium cause spectral line shifts of the impurity atoms which can exceed the natural linewidth. These shifts depend mainly on the type of chemical bond of a dielectric, the symmetry of the internal fields and the temperature.

In many practical applications the efficiency of impurity centers can be improved by increasing of their density. For dense ensembles, in which the average distance between impurity centers is comparable with the wavelength of resonant radiation, the dipole-dipole interaction between themselves plays an important role [25] – [26]. Cooperative effects cause density-dependent shifts of atomic transition as well as distortion of spectral line shape [27]. This influences both on the properties of atoms [28] – [30] as well as on scattered radiation characteristics [31] – [36].
It is important that the radiative transfer between different atoms located inside the cavity is essentially modified compared with absence of a cavity [37]. Today the interatomic interaction in the atomic systems coupled to a nanofiber [38] as well as to phonic crystals [39] – [41] is intensively investigated. Nevertheless, multiatomic cooperative effects inside the cavity, including multiple and recurrent scattering have not yet been studied in detail. It is connected with both theoretical problems as well as some computational difficulties.

In this paper we study a disordered ensemble, which consists of $N \gg 1$ impurity centers in a Fabry-Perot cavity taking into account the transition frequency shifts of impurity centers caused by the internal fields of a dielectric medium. On the basis of the general theory we study the spontaneous decay dynamics of the local atomic excitation prepared inside a cavity depending on the distance between the mirrors. In particular, we analyze the case when the distance between two mirrors $d$ is less than a half of the transition wavelength $\lambda_0$ as well as the case when $d \gg \lambda_0$. This first case attracts a special attention due to the fact that the spontaneous decay from some Zeeman sublevels of atomic exited state is suppressed. This indicates that the use of a cavity allows to achieve very large time of radiation trapping, much more than in the case of absence of a cavity [42].

2. Basic assumptions and approach

Let us consider a disordered ensemble, which consists of $N$ motionless impurity atoms in a transparent dielectric closed in a Fabry-Perot cavity. The mirrors of a cavity are assumed to be perfectly conducting. The sizes of the mirrors are much larger than resonant transition wavelength $\lambda_0$, the distance between the mirrors $d$ and the average distance between impurity atoms (see Fig. 1). It allows us to consider the sizes of the mirrors equal to infinity.

![Figure 1. Sketch of the cavity and the atomic ensemble.](image)

We assume that the temperature is low enough to neglect the interaction of impurity atoms with phonon reservoir. It allows us to assume the quantum state of the joint system, which consists of atoms and electromagnetic field to be pure, and describe it by the wave function.

Even for a transparent dielectric the transition frequency of impurity atoms in a dielectric $\omega_a$ can be different from the transition frequency of a free atom $\omega_0$. It is connected with internal
fields of a dielectric. Thus, \( \omega_a = \omega_0 + \Delta_a \), where \( \Delta_a \) is the frequency shift of the atom \( a \) which depends on its spatial position, \( a = 1, \ldots, N \).

In this paper we use quantum microscopic approach described firstly in [43] and developed afterward in [31] for description of collective effects in dense and cold nondegenerate atomic gases. It was earlier successfully used for analysis of optical properties of dense atomic ensembles [28] – [30] as well as for studying of light scattering from such ensembles [34] – [42].

This approach is based on solution of the non-stationary Schrodinger equation for the wave function \( \psi \) of the joint system consisting of atoms and the electromagnetic field, including vacuum reservoir

\[
i \hbar \frac{\partial \psi}{\partial t} = \hat{H} \psi. \tag{1}
\]

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

\[
\hat{H} = \hat{H}_0 + \hat{V},
\]

\[
\hat{H}_0 = \hat{H}_f + \sum_a \hat{H}_a. \tag{2}
\]

In the dipole approximation used here, we have

\[
\hat{V} = -\sum_a \hat{d}^{(a)} \hat{E}(\mathbf{r}_a). \tag{3}
\]

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

The electric field operator \( \hat{E}(\mathbf{r}) \) in a cavity can be obtained in a standard way by quantization of the classical field \( E(\mathbf{r}, t) \). The latter is a solution of Maxwell equations with corresponding boundary conditions. Let us consider \( z \) axis perpendicular to the mirrors, \( x \) and \( y \) axis parallel to the mirrors (see Fig. 1). \( z = 0 \) corresponds to the position of the first mirror and \( z = d \) to the second mirror. The boundary conditions can be written as follows: \( E_x|_{z=0} = E_x|_{z=d} = E_y|_{z=0} = E_y|_{z=d} = 0 \). In the considered geometry we have

\[
\hat{E}(\mathbf{r}) = \sum_{k,\alpha} \frac{i \omega_k}{c} \sqrt{\frac{\hbar}{2 \omega_k}} \hat{a}_{k,\alpha} B_{k,\alpha}^{0} \{ e_x u_{k,\alpha}^x \sin(k_n z) + e_y u_{k,\alpha}^y \sin(k_n z) + e_z u_{k,\alpha}^z \cos(k_n z) \} \exp(i\mathbf{k} \cdot \mathbf{r}_a) + \text{H.c.} \tag{4}
\]

In this equation \( B_{k,\alpha}^{0} \) is the normalization factor, given by

\[
B_{k,\alpha}^{0} = \sqrt{\frac{8 \pi c^2}{L^2 d}} \times \begin{cases} 1, & \text{if } n \in N \\ 1/\sqrt{2}, & \text{if } n = 0. \end{cases} \tag{5}
\]

\( L \) is the longitudinal size of the quantization volume.

We will seek the wave function \( \psi \) as an expansion in a set of eigenfunctions \( \psi_l \) of the operator \( H_0 \):

\[
\psi = \sum_l b_l(t) \psi_l. \tag{6}
\]
Here, the subscript $l$ defines the state of all atoms and the field. Using this representation of the wave function we convert the equation (1) to the system of linear differential equations for the quantum amplitudes of states

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

(8)

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

The total number of equations in the system (8) is equal to infinity. It is caused by the infinity number of the degrees of freedom of electromagnetic field.

The key simplification of the approach employed is in restriction of the total number of states $|l|$ 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 [44]):

1. One-fold atomic excited states

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

2. Resonant single-photon states

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

3. Nonresonant states with two excited atoms and one photon

   $$\psi_{e_a e_b} = |g, ..., g, e, g, ..., g, e, g, ..., g \rangle \otimes |k, \alpha\rangle,$$

   $$E_{e_a e_b} = \hbar (\omega_{a} + \omega_{b}) + \hbar \omega_{k}.$$

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 dipole-dipole interaction at short interatomic distances, comparable with $\lambda_0$.

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 atomic and field subsystem

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

In the framework of the assumptions considered here, the quantum amplitude of the state $\psi_{g'}$ does not change during the evolution of the considered system. It is explained by the fact that, any transitions between $\psi_{g'}$ and other quantum states taken into account are impossible. The Lamb shift is considered to be included in $\omega_0$.

Despite the restriction of the total number of quantum states, the set of equations remains infinite because it contains the field states. We can, however, exclude amplitudes of states with one photon and obtain a finite closed system of equations for the atomic states $b_e$. For Fourier components $b_e(\omega)$ we have (at greater length; see [31])

$$\sum_{e'} \left[ (\omega - \omega_{a}) \delta_{ee'} - \sum_{ee'} (\omega) \right] b_{e'}(\omega) = i \delta_{e0}.$$

(9)

This 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 their ground state at $t = 0$ and electromagnetic field is in the vacuum state. The system (9) with the initial conditions considered here allows us to analyze both stationary light scattering as well as nonstationary problems. In particular, the time dependent excitation probability of initially excited atom $P_{g}(t) = |b_{o}(t)|^2$ can be calculated.

The size of the system (9) is determined by the number of atoms $N$ and the structure of the energy levels. In this paper we consider that the ground state is characterized by the total angular momentum $J = 0$ and the excited state $-J = 1$. Note that, in the case considered, there are three excited sublevels for each atom $e = |J, m\rangle$, which differ by the value of angular momentum projection $m = -1, 0, 1$. Therefore, the total number of one-fold atomic excited
states is $3N$. This scheme of levels corresponds to atoms with 2 valence electrons such as Sr, Yb, Ca.

The matrix $\Sigma_{ee}(\omega)$ describes both spontaneous decay and photon exchange between the atoms. It plays a key role in the microscopic theory. The explicit expression for this matrix corresponding to the cavity was derived in Refs. [45] – [46]. It extremely differs from one corresponding to the free space. This is explained by the fact, that a cavity modifies the structure of modes of electromagnetic field.

In the next section, we will use the general approach to investigate the simultaneous influence of the cavity and interatomic dipole-dipole interaction on the spontaneous decay dynamics of an excited atom depending on the distance between two mirrors.

3. Results and discussion

The inverse Fourier transform of $b_\nu(\omega)$ allows us to obtain the time dependence of the quantum amplitudes of the one-fold atomic excited states $b_\nu(t)$. The spontaneous decay dynamics of initially excited atom is determined by $P_\nu(t) = |b_\nu(t)|^2$.

The influence of the dipole-dipole interaction on the properties of atomic ensemble is determined not only by the density of impurity atoms. 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. In the present paper we restrict ourselves by the assumption 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 $\Delta_\nu$ can be presented as a sum of its average value $\Delta$ 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 r.m.s. deviation $\delta$. In this paper we will restrict ourselves by the case when $\Delta$ is the same for all Zeeman sublevels of the excited state. This corresponds to the cubic symmetry of internal fields of a medium, for instance. Hereafter we will consider $\Delta$ to be included in the resonant transition frequency $\omega_0$. The ratio of $\delta$ to natural linewidth of the atoms $\gamma_0$ characterizes the degree of resonance between impurity atoms. This is one of a key parameters of the considered system in our theory. In particular, it can be considered the case when all atoms are resonant to each other $\delta \ll \gamma_0$ as well as opposite case when inhomogeneous broadening is much larger than natural linewidth $\delta \gg \gamma_0$. The criterion of the significance of cooperative effects for the last case was derived in Refs. [45] – [46]. It can be written as follows: $(n\gamma_0/\delta)^{-1/3} \leq \lambda_0$. This criterion can be generalized on the atomic media with inhomogeneous broadening of different nature. In particular, it can be used for the analysis of the gas cells with alkali atoms at room temperature that play a key role in frequency standards based on CPT [47] – [49].

In this paper we will focus our attention on the investigation of the cooperative spontaneous decay of an excited atom depending on the distance between the mirrors. Let us consider initially excited atom located in the middle between the mirrors $z_{exc} = d/2$ (reference point $z = 0$ corresponds to the left mirror) and in the center of atomic ensemble. All other atoms are considered to be in their ground states at initial time $t = 0$ and all field modes in the vacuum state.

Fig. 2 shows the spontaneous decay dynamics from Zeeman sublevel $m = -1$ or $m = 1$ of the initially excited central atom. Atomic density is chosen $n = 0.05$ (in this paper we consider $k_0^{-1} = c/\omega_0$ as a unit of length). Cooperative effects play an important role in the ensembles of such density even in the case of absence of a cavity [27]. Comparing the curves in Fig. 2 we conclude that for small distance between the mirrors the decay dynamics in a cavity essentially differs from the case of absence of a cavity. However, as $d$ increases the cooperative decay in a cavity gradually tends to the case of absence of a cavity.

Fig. 3 shows the spontaneous decay dynamics from Zeeman sublevel $m = 0$. We can conclude
that for the parameters considered here the spontaneous decay in a cavity with \( d > 10 \) is almost the same as without a cavity. All results shown here are averaged over random spatial positions of impurity atoms. Very small fluctuations on the curves demonstrate the accuracy of calculations.

4. Conclusion

We have studied the cooperative spontaneous decay of an excited impurity atom embedded in a transparent dielectric and located in a Fabry-Perot cavity. It was performed on the basis of the quantum microscopic approach developed in Refs. [45] – [46]. In this paper the main attention was given to the investigation of the character of the cooperative effects depending on the distance \( d \) between the mirrors of a cavity. It was shown that in the case \( d \leq \lambda_0 \) cooperative spontaneous decay in a cavity essentially differs from one without a cavity and for \( d > \lambda_0 \) it is almost the same. In particular, it was demonstrated that for the atomic density \( n = 0.05 \) and inhomogeneous broadening \( \delta \ll \gamma_0 \) the discrepancy in the spontaneous decay in a cavity and without a cavity can be neglected if \( d > 10 \).

In our opinion, the approach used in the present paper allows us to investigate the influence of a cavity on the observability 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 [50] – [51].

The theory discussed here can be generalized to the case of the real refractive index of the dielectric. In addition, it can be further generalized to the atomic ensembles in the waveguide. The last attracts particular interest due to spontaneous decay suppression of all the Zeeman sublevels of the atomic excited state under the condition when the resonant frequency of atomic transition is less than the cut-off frequency of the waveguide.

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