

Coherent effects under suppressed spontaneous emission

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Abstract. An ensemble of resonance atoms is considered, which are doped into a medium with well developed polariton effect, when in the spectrum of polariton states there is a band gap. If an atom with a resonance frequency inside the polariton gap is placed into the medium, the atomic spontaneous emission is suppressed. However, a system of resonance atoms inside the polariton gap can radiate when their coherent interaction is sufficiently strong. Thus the suppression of spontaneous emission for a single atom can be overcome by a collective of atoms radiating coherently. Conditions when such collective effects can appear and their dynamics are analysed.

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1 Introduction

Inhibition of spontaneous emission has first been predicted [1,2] to occur in three-dimensional periodic structures, in which, due to periodicity, an electromagnetic band gap develops. As a result, the spontaneous emission with a frequency inside the band gap is rigorously forbidden. This type of matter, where photon band gap arises because of the structural periodicity in real space, has been called photonic band-gap materials [3].

Band gaps also appear in natural dense media due to photon interactions with optical collective excitations, such as phonons, magnons, excitons, and so on [4,5]. This kind of a gap is termed the polariton band gap. If a single atom is placed in a medium with the polariton band gap, and the atomic resonance frequency lies inside the gap, then the atom spontaneous emission can be suppressed [6,7].

Physical reasons for the effect of suppressed spontaneous emission are rather similar for both types of materials, the difference being in the nature of scatterers light interacts with. In artificial photonic band-gap materials, a suppression of the photon density of states over a narrow frequency range results from multiple photon scattering by spatially correlated scatterers. In natural dense media, such as dielectrics or semiconductors, a frequency gap for electromagnetic modes develops as a result of photon scattering by optical collective excitations. The suppression of spontaneous emission can be explained as follows. Let us imagine an excited atom in a medium, with the atomic transition frequency within the prohibited gap. The atom

tends to become deexcited by emitting a photon. However, since the propagation of photons inside the gap is prohibited, the emitted photon is scattered back and is again absorbed by the atom. This means that the atom cannot become deexcited or, in other words, that spontaneous emission is suppressed. Hence, this effect is characterized by the condition

$$\lim_{t \rightarrow \infty} s(t) > -1$$

for the average population difference $s \equiv \langle \sigma_i^z \rangle$ that is a statistical average of the Pauli operator σ_i^z . In the described physical picture, the limit $t \rightarrow \infty$ is, of course, conditional simply implying that the suppression of spontaneous emission happens during times much longer than the emission time γ^{-1} , where γ is the spontaneous emission rate. The complete suppression for infinite times is, certainly, never possible since there always exist spontaneous photons with frequencies out of the polariton band gap, so that sooner or later an atom becomes deexcited *via* nonresonant photons. However, for sufficiently large polariton gaps, such nonresonant relaxation processes are very slow. The characteristic relaxation time of a nonresonant process occurring through a frequency gap Δ can be estimated [8,9] as $\gamma^{-1}(1 + \Delta^2/\gamma^2)$. Accepting the values typical of the majority of semiconductors [4,5,10–12], for the polariton gap one has $\Delta \sim 10^{13} \text{ s}^{-1}$ while the emission rate is $\gamma \sim 10^8\text{--}10^9 \text{ s}^{-1}$. Hence the characteristic time of relaxation through nonresonant processes is 0.1–100 s, which is essentially longer than typical emission times of free atoms. In this paper, we do not take into account the nonresonant deexcitation of atoms, which is justified for rather long times as estimated above.

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Even though spontaneous emission of a single atom in a medium is suppressed, the situation for an ensemble of atoms can be different. When a collective of resonance atoms is doped into the medium with a band gap, an impurity band can be formed within the prohibited gap. This happens in photonic band-gap materials [13] as well as in media with the polariton gap [14,15]. For the collection of identical resonance atoms with spacing much shorter than the transition wavelength, electromagnetic effective interaction takes place due to photon exchange. For closely spaced atoms with transition frequencies in the gap, the virtual photon exchanges are of such energy that the atoms do not experience the existence of the gap [16, 17]. When an impurity band is formed, electromagnetic radiation becomes possible. Sufficiently strong effective interactions collectivise the resonance atoms that start radiating coherently. Thus, the suppression of spontaneous emission for a single atom can be overcome by a collective of atoms. Hence the suppressed light can be liberated, at least partially. But what would be the dynamics of such a collective radiation?

In the case of photonic band-gap materials, spontaneous emission near the edge of a photonic band-gap has been considered [18,19] for a point-like model with the radiation wavelength much larger than the model linear size, $\lambda \gg L$. For polariton-gap media, the dynamics of collective liberation has not yet been properly studied. Moreover, the consideration of point-like models, with $\lambda \gg L$, in the case of suppressed spontaneous emission cannot be accepted as realistic because of the follows. Assume that an excited atom, with a transition frequency within the prohibited photonic gap, is placed into a bandgap material. A photon emitted by the atom, before being reflected back, travels at the distance called the localization length l_{loc} that is about several wavelengths, $l_{\text{loc}} > \lambda$. It is only when the localization length is much shorter than the sample size, $l_{\text{loc}} \ll L$, there is sense of talking that light can be localized, hence spontaneous emission be suppressed. While for a concentrated point-like system, with $\lambda \gg L$, one has even more $l_{\text{loc}} > \lambda \gg L$. That is, an emitted photon will safely quit the sample, being never reflected back. Therefore the localization of light and suppressed emission principally cannot occur in a point-like system.

In the present paper, we consider a system of N resonance atoms doped into a polariton-gap medium, in which spontaneous emission of a single atom is suppressed. The general case of arbitrary wavelengths is studied, which makes it necessary to take into account local-field effects. The dynamics of radiation is carefully analysed. Two basic points difference the present consideration from the works of other authors: first, a realistic situation is analysed, when the radiation wavelength can be (and *must be*, for the spontaneous emission be suppressed and light be localized) much shorter than the sample size. Second, the transition frequency is assumed to lie not at the edge of the gap but *deeply inside* the prohibited bandgap. The system of units is used with the Planck constant $\hbar \equiv 1$.

2 Evolution equations

Aiming at studying the dynamics of more or less realistic system, we start with the Hamiltonian

$$\hat{H} = \hat{H}_a + \hat{H}_f + \hat{H}_{af} + \hat{H}_m + \hat{H}_{mf} \quad (1)$$

containing the following parts. The Hamiltonian of two-level resonance atoms

$$\hat{H}_a = \frac{1}{2} \sum_{i=1}^N \omega_0 (1 + \sigma_i^z), \quad (2)$$

with the transition frequency ω_0 . The radiation-field Hamiltonian

$$\hat{H}_f = \frac{1}{8\pi} \int (\mathbf{E}^2 + \mathbf{H}^2) \, \text{dr}, \quad (3)$$

with electric field \mathbf{E} and magnetic field $\mathbf{H} = \nabla \times \mathbf{A}$. The vector potential \mathbf{A} is assumed to satisfy the Coulomb gauge calibration $\nabla \cdot \mathbf{A} = 0$ and the commutation relation

$$[E^\alpha(\mathbf{r}, t), A^\beta(\mathbf{r}', t)] = 4\pi ic \delta_{\alpha\beta} \delta(\mathbf{r} - \mathbf{r}'),$$

where c is the light velocity. The atom-field interaction is given by the standard dipole Hamiltonian

$$\hat{H}_{af} = - \sum_{i=1}^N \left(\frac{1}{c} \mathbf{j}_i \cdot \mathbf{A}_i + \mathbf{d}_i \cdot \mathbf{E}_{0i} \right), \quad (4)$$

in which $\mathbf{A}_i \equiv \mathbf{A}(\mathbf{r}_i, t)$; the transition-current and the transition-dipole operators, respectively, are

$$\mathbf{j}_i = i\omega_0 (\mathbf{d}\sigma_i^+ - \mathbf{d}^*\sigma_i^-), \quad \mathbf{d}_i = \mathbf{d}\sigma_i^+ + \mathbf{d}^*\sigma_i^-, \quad (5)$$

where \mathbf{d} is the transition dipole and σ_i^\pm are the ladder operators; and \mathbf{E}_{0i} is an external field, if any. The Hamiltonian of matter, \hat{H}_m , can be modelled by a collection of oscillators [6,7]. And matter-field interactions are given by the Hamiltonian

$$\hat{H}_{mf} = - \frac{1}{c} \sum_{i=1}^{N_0} \mathbf{j}_{mi} \cdot \mathbf{A}_i, \quad (6)$$

where \mathbf{j}_{mi} is a current produced by matter at the point \mathbf{r}_i ; N_0 being the number of oscillators modelling the matter.

When considering stationary properties of electromagnetic field, one usually passes to the momentum-energy representation by expanding operators in Fourier series. However this representation is not as convenient for studying dynamical properties of a system of atoms, especially when their transition wavelength is much shorter than the system size. Therefore we shall employ here the space-time representation whose brief description can be found in book [20] and detailed analysis in references [21,22].

The Heisenberg equations of motion, with the Hamiltonian (1), yield for the atomic variables the equations

$$\begin{aligned} \frac{d\sigma_i^-}{dt} &= -i\omega_0\sigma_i^- + (k_0\mathbf{d} \cdot \mathbf{A}_i - i\mathbf{d} \cdot \mathbf{E}_{0i})\sigma_i^z, \\ \frac{d\sigma_i^z}{dt} &= -2(k_0\mathbf{d} \cdot \mathbf{A}_i - i\mathbf{d} \cdot \mathbf{E}_{0i})\sigma_i^+ \\ &\quad - 2(k_0\mathbf{d}^* \cdot \mathbf{A}_i + i\mathbf{d}^* \cdot \mathbf{E}_{0i})\sigma_i^-, \end{aligned} \quad (7)$$

where $k_0 \equiv \omega_0/c$. The Heisenberg equations for the electric field and vector potential lead to the Maxwell operator equations, combining which with the Coulomb gauge calibration, one gets

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \mathbf{A} = -\frac{4\pi}{c} \mathbf{J}, \quad (8)$$

with the density of current

$$\mathbf{J}(\mathbf{r}, t) = \sum_{i=1}^N \mathbf{j}_i(t) \delta(\mathbf{r} - \mathbf{r}_i) + \sum_{i=1}^{N_0} \mathbf{j}_{mi}(t) \delta(\mathbf{r} - \mathbf{r}_i). \quad (9)$$

The formal operator solution to equation (8) reads

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}_{\text{vac}}(\mathbf{r}, t) + \frac{1}{c} \int \mathbf{J} \left(\mathbf{r}', t - \frac{1}{c} |\mathbf{r} - \mathbf{r}'| \right) \frac{d\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|}, \quad (10)$$

where \mathbf{A}_{vac} is a solution of the related uniform equation. Substituting the density of current (9) into equation (10) results in the vector potential

$$\mathbf{A} = \mathbf{A}_{\text{vac}} + \mathbf{A}_{\text{rad}} + \mathbf{A}_{\text{mat}} \quad (11)$$

consisting of the potential \mathbf{A}_{vac} of vacuum fluctuations, potential \mathbf{A}_{rad} produced by radiating atoms, and the potential \mathbf{A}_{mat} induced by the medium,

$$\mathbf{A}_{\text{rad}}(\mathbf{r}_i, t) = \sum_j^N \frac{1}{c r_{ij}} \mathbf{j}_j \left(t - \frac{r_{ij}}{c} \right),$$

$$\mathbf{A}_{\text{mat}}(\mathbf{r}_i, t) = \sum_j^{N_0} \frac{1}{c r_{ij}} \mathbf{j}_{mj} \left(t - \frac{r_{ij}}{c} \right), \quad (12)$$

where the summation excludes the terms with $j = i$, and the notation $r_{ij} \equiv |\mathbf{r}_{ij}|$, $\mathbf{r}_{ij} \equiv \mathbf{r}_i - \mathbf{r}_j$, is used. It is worth emphasizing that the relation (10) is an exact expression immediately following from the evolution equation (8) and identical to the latter. No far-zone approximation is involved here. This approximation could be done [23] by expanding $|\mathbf{r} - \mathbf{r}'|^{-1}$ in the integrand of equation (10) in powers of r'/r , assuming that $r \equiv |\mathbf{r}| \gg r' \equiv |\mathbf{r}'|$. But we do not invoke here such an expansion. Substituting equations (10–12) into the Hamiltonian (4) and the evolution equations (7), it is easy to notice that all these equations contain an effective dipole-dipole coupling of atoms responsible for near-zone local-field effects [24]. Expression (10) is not a final solution for the observable field but it is an *operator relation* having sense only in the combination with the Heisenberg equations (7). The system of equations (7, 10) follows directly from the initial Hamiltonian (1) and is often employed in optics [20].

Let us introduce the quantities

$$u_i(t) \equiv \langle \sigma_i^-(t) \rangle, \quad s_i(t) \equiv \langle \sigma_i^z(t) \rangle, \quad (13)$$

in which the angle brackets imply the statistical averaging over the *atomic degrees of freedom* only, not touching the

vacuum and matter degrees of freedom. For the double correlators, we shall employ the mean-field-type decoupling

$$\langle \sigma_i^\alpha \sigma_j^\beta \rangle = \langle \sigma_i^\alpha \rangle \langle \sigma_j^\beta \rangle \quad (i \neq j). \quad (14)$$

It is worth emphasizing that, since only the atomic degrees of freedom have been involved in the averaging, the decoupling (14) does not kill quantum effects related to the vacuum and the matter degrees of freedom, which have not yet been averaged out. The variables corresponding to these degrees of freedom will enter the equations of motion for the quantities (13) as operator variables or they can be modelled by stochastic variables [25]. This is why the decoupling (14) may be called the *stochastic mean-field approximation*.

Another problem concerns the way of treating the retardation appearing in equations (12). We cannot use the Markov approximation just ignoring this retardation since this would eliminate all local-field effects that are so important for realistic many-atom systems with wavelength shorter than the system size [26]. But we may employ the Born approximation

$$\left\langle \sigma_j^- \left(t - \frac{r_{ij}}{c} \right) \right\rangle = u_j(t) \exp(ik_0 r_{ij}) \quad (15)$$

retaining the account of local fields. Recall that considering resonance atoms, one always keeps in mind that the intensities of interactions of atoms with all fields are assumed to be much smaller than the transition frequency ω_0 . If this were not so, then the notion of resonance atoms as such would lose its sense, because strong fields, essentially shifting the atomic energy levels, would completely change the classification of these levels and the values of transition frequencies. Under the assumption that the transition frequency ω_0 is the largest energy scale, from equations (7) it immediately follows that the approximation (15) is completely justified [20–22]. This can also be called the quasirelativistic approximation since in the non-relativistic limit, when $c \rightarrow \infty$, equation (15) becomes an identity.

The equations for the quantities (13) follow from equations (7) where we introduce the notation for the effective field

$$f_i(t) \equiv k_0 \langle \mathbf{d} \cdot \mathbf{A}_{\text{rad}}(\mathbf{r}_i, t) \rangle - i \mathbf{d} \cdot \mathbf{E}_{0i} + \xi_i(t), \quad (16)$$

in which the last term is the random field

$$\xi_i(t) \equiv k_0 \mathbf{d} \cdot [\mathbf{A}_{\text{vac}}(\mathbf{r}_i, t) + \mathbf{A}_{\text{mat}}(\mathbf{r}_i, t)]. \quad (17)$$

As usual, we also need to take into account the level width γ_1 and the line width γ_2 . The introduction of these widths is done in the standard way [23, 25, 27] keeping in mind that the values of γ_1 and γ_2 , in general, differ from the emission rate γ of a free atom. This is because the longitudinal relaxation rate γ_1 is assumed to effectively include the influence of matter which atoms are doped in, for instance, the influence of lattice vibrations. The transverse relaxation rate γ_2 differs from γ because of the same reason of incorporating the influence of matter, for example,

by taking account of dynamic and, partially, of inhomogeneous broadening. Thus, γ_1 and γ_2 are to be treated as phenomenological parameters introduced in the commonly accepted way [23,25,27]. In this way, we obtain the equations for the average transition amplitude

$$\frac{du_i}{dt} = -(i\omega_0 + \gamma_2)u_i + f_i s_i, \quad (18)$$

and the average population difference

$$\frac{ds_i}{dt} = -2(u_i^* f_i + f_i^* u_i) - \gamma_1(s_i - s_0). \quad (19)$$

The last term in equation (19) allows for the suppression of spontaneous emission in the case of a single atom. Really, to return to the latter case, we have to put $f_i = 0$. Then we get the equation for a single atom, with the solution $s_i(t) = s_0$, as is discussed in the Introduction. But, in the case of an ensemble of atoms, the presence of nonlinear terms related to the effective field (16) can make the solutions to the evolution equations quite nontrivial. These equations are to be complemented by an equation either for u_i^* or for $|u_i|^2 \equiv u_i^* u_i$. For the latter equation, we find

$$\frac{d|u_i|^2}{dt} = -2\gamma_2|u_i|^2 + s_i(u_i^* f_i + f_i^* u_i). \quad (20)$$

Equations (18–20) are the main evolution equations we shall consider. These are the stochastic differential equations since they contain the random field (17) including the vacuum and matter degrees of freedom. Electromagnetic fluctuations coupled with collective excitations of a medium are called polaritons [4,5]. Therefore equation (17) represents the polariton field acting on atoms.

Strictly speaking, one should distinguish two different physical cases, depending on the relation between the longitudinal relaxation time $T_1 \equiv \gamma_1^{-1}$ and the characteristic time of nonresonant processes $T_n = \gamma^{-1}(1 + \Delta^2/\gamma^2)$ discussed in the introduction. As is explained in the latter, it makes sense of talking about the suppressed spontaneous emission of an atom only for times $t \ll T_n$. This suppression can be either dynamic, when $T_1 \ll T_n$, or static, when $T_1 \sim T_n$. In the first case, one has to retain the parameter γ_1 in the evolution equations, while in the second case, one has to set $\gamma_1 = 0$. For generality, we shall consider both these situations starting with the more general case of finite γ_1 , from which it is easy to pass to the particular case of $\gamma_1 = 0$.

3 Scale separation

In what follows, we consider the case when an external field is used only for exciting atoms at the initial time, but after this the field is switched off, $\mathbf{E}_{0i} = 0$. It is worth saying a few words about the possibility of exciting resonance atoms whose transition frequency lies in the polariton band gap. There can be several ways of doing this. The first way is by exciting the atoms with a short pulse

of a resonance external field. The amplitude of the latter is to be sufficiently large since a weak electromagnetic wave with a frequency within the polariton gap cannot propagate through the medium because of total reflection. However, when the incident field is strong enough, a monochromatic wave can penetrate into the medium with a polariton gap due to nonlinear effects [28,29]. Another possibility for exciting the atoms in a polariton gap could be through a third level with the related transition frequency lying outside the gap. It could also be possible to get the population inversion at the desired level through a set of levels outside the gap, by using nonresonance pumping.

It is convenient to define the arithmetic averages

$$u(t) \equiv \frac{1}{N} \sum_{i=1}^N u_i(t), \quad s(t) \equiv \frac{1}{N} \sum_{i=1}^N s_i(t) \quad (21)$$

of the variables (13) and also the average polariton field

$$\xi(t) \equiv \frac{1}{N} \sum_{i=1}^N \xi_i(t). \quad (22)$$

Let us introduce the collective frequency Ω and the collective width Γ , respectively,

$$\Omega \equiv \omega_0 + \gamma_2 g' s, \quad \Gamma \equiv \gamma_2(1 - g s), \quad (23)$$

where the effective atomic coupling parameters are

$$g \equiv \frac{3\gamma}{4\gamma_2 N} \sum_{i \neq j}^N \frac{\sin k_0 r_{ij}}{k_0 r_{ij}}, \quad g' \equiv \frac{3\gamma}{4\gamma_2 N} \sum_{i \neq j}^N \frac{\cos k_0 r_{ij}}{k_0 r_{ij}}, \quad (24)$$

and the notation

$$\gamma \equiv \frac{4}{3} k_0^3 d_0^2, \quad k_0 \equiv \frac{\omega_0}{c}, \quad \mathbf{d} \equiv d_0 \mathbf{e}_d$$

is employed. Then from equations (18–20) in the mean-field approximation, we obtain

$$\frac{du}{dt} = -(i\Omega + \Gamma)u + s\xi + \gamma_2(g + ig')su^* \mathbf{e}_d^2, \quad (25)$$

$$\begin{aligned} \frac{ds}{dt} = & -4\gamma_2 g |u|^2 - 2(u^* \xi + \xi^* u) - \gamma_1(s - s_0) \\ & - 2\gamma_2 [(g + ig')(u^* \mathbf{e}_d)^2 + (g - ig')(\mathbf{e}_d^* u)^2], \end{aligned} \quad (26)$$

$$\begin{aligned} \frac{d|u|^2}{dt} = & -2\Gamma|u|^2 + s(u^* \xi + \xi^* u) \\ & + \gamma_2 s [(g + ig')(u^* \mathbf{e}_d)^2 + (g - ig')(\mathbf{e}_d^* u)^2]. \end{aligned} \quad (27)$$

Equations (25–27) form a system of nonlinear stochastic differential equations. It is worth stressing that the rotating wave approximation (RWA), well-known in resonance optics [20,23,25,27], cannot be blindly applied for simplifying this system of equations. Really, the standard usage of RWA is as follows. One assumes that the polarization variable u behaves approximately as $\exp(-i\Omega t)$,

that is, rotates with the frequency Ω , while the population difference s does not rotate. With this assumption, one neglects in the right-hand sides of the evolution equations the terms that behave qualitatively differently to the assumed behaviour of the left-hand sides. Thus, in equation (25) one should drop the counter-rotating term containing u^* as well as the non-rotating term $s\xi$. Similarly, in equations (26, 27) for non-rotating quantities, one should neglect the rotating terms with u^2 and $(u^*)^2$ as well as with $u\xi^*$ and ξ^*u . But, as is evident, such a procedure would completely lose information of polariton degrees of freedom ξ . Hence, RWA is not applicable to such stochastic differential equations. Although the RWA reasoning is not justified for these equations, one may try to simplify the latter by formally omitting only those terms that would be dropped if the polariton field ξ were absent, but leaving untouched the terms containing ξ . Then one would come to the system of equations

$$\begin{aligned}\frac{du}{dt} &= -(i\Omega + \Gamma)u + s\xi, \\ \frac{ds}{dt} &= -4\gamma_2g|u|^2 - 2(u^*\xi + \xi^*u) - \gamma_1(s - s_0), \\ \frac{d|u|^2}{dt} &= -2\Gamma|u|^2 + s(u^*\xi + \xi^*u).\end{aligned}$$

This is yet a system of nonlinear, because of equations (23), stochastic differential equations.

To solve this system of equations (25–27), we shall use the scale separation approach [30–32] which is a variant of the Krylov-Bogolubov averaging method [33] generalized to the case of stochastic differential equations. To this end, we need, first of all, to classify the functional variables of the system (25–27) onto fast and slow. This can be done by accepting the usual inequalities

$$\frac{\gamma_1}{\Omega} \ll 1, \quad \frac{\gamma_2}{\Omega} \ll 1, \quad \left| \frac{\Gamma}{\Omega} \right| \ll 1 \quad (28)$$

and assuming that the average energy of atomic interactions with the polariton field is also much less than Ω . Then from equations (25–27) it immediately follows that the variable u is to be classified as fast as compared to the slow variables s and $|u|^2$. The next step is to solve equation (25) for the fast variable treating the slow variables as quasi-invariants, which yields

$$u(t) = \left[u_0 + s \int_0^t e^{(i\Omega + \Gamma)t'} \xi(t') dt' \right] e^{-(i\Omega + \Gamma)t}, \quad (29)$$

where $u_0 \equiv u(0)$. This solution is to be substituted into the right-hand sides of the equations (26, 27) for the slow variables, with averaging these right-hand sides over polariton degrees of freedom and over explicitly entering time. In accomplishing this procedure, we define the effective atom-polariton coupling

$$\alpha \equiv \frac{\text{Re}}{s\Gamma} \lim_{\tau \rightarrow \infty} \frac{1}{\tau} \int_0^\tau \langle\langle u^*(t)\xi(t) \rangle\rangle dt, \quad (30)$$

where Re means the real part and $\langle\langle \dots \rangle\rangle$ implies the averaging over polariton degrees of freedom. With the form (29) and the condition $\langle\langle \xi(t) \rangle\rangle = 0$, this gives

$$\alpha = \frac{\text{Re}}{\Gamma} \lim_{\tau \rightarrow \infty} \frac{1}{\tau} \int_0^\tau dt \int_0^t e^{-(i\Omega + \Gamma)(t-t')} \langle\langle \xi^*(t)\xi(t') \rangle\rangle dt'.$$

By assumption, the atom-polariton coupling is weak,

$$|\alpha| \ll 1. \quad (31)$$

In order to understand the structure of the coupling (30), let us model the polariton field by an ensemble of N_0 oscillators, so that

$$\xi(t) = \frac{1}{\sqrt{N_0}} \sum_k \gamma_k \left(b_k e^{-i\omega_k t} + b_k^\dagger e^{i\omega_k t} \right), \quad (32)$$

where b_k and b_k^\dagger are Bose operators with the properties

$$\begin{aligned}\langle\langle b_k^\dagger b_{k'} \rangle\rangle &= \delta_{kk'} n_k, & \langle\langle b_k b_{k'} \rangle\rangle &= 0, \\ \langle\langle b_k b_{k'}^\dagger \rangle\rangle &= \delta_{kk'} (1 + n_k), & \frac{1}{N_0} \sum_k n_k &= 1,\end{aligned}$$

and the effective width γ_k is of order γ outside the polariton gap but is zero inside this gap,

$$\gamma_k = 0, \quad \omega_1 < \omega_k < \omega_2, \quad \Delta_p \equiv \omega_2 - \omega_1.$$

For the polariton field (32), the coupling (30) becomes

$$\alpha = \frac{1}{N_0} \sum_k |\gamma_k|^2 \left[\frac{n_k}{(\omega_k - \Omega)^2 + \Gamma^2} + \frac{1 + n_k}{(\omega_k + \Omega)^2 + \Gamma^2} \right]. \quad (33)$$

We assume that the collective atomic frequency Ω lies *deeply inside* the polariton gap of width Δ_p . If this width is much smaller than the frequencies at the edges of the gap, $\Delta_p \ll \omega_1$, then equation (33) can be approximated by the form

$$\alpha \approx \frac{4\gamma^2}{\Delta_p^2 + 4\Gamma^2}.$$

After substituting equation (29) into the right-hand sides of equations (26, 27), with accomplishing the described averaging and introducing the new function

$$w \equiv \langle\langle |u|^2 \rangle\rangle - \alpha s^2, \quad (34)$$

we obtain the equations

$$\frac{ds}{dt} = -4g\gamma_2 w - \gamma_1(s - s_0), \quad \frac{dw}{dt} = -2\gamma_2(1 - gs)w \quad (35)$$

for the slow variables.

Let us note that the system of three equations (25–27) has been derived from the set of $3N$ equations (18–20), where $i = 1, 2, \dots, N$, by employing a mean-field type approximation for the averages (21). As is obvious, it is impossible to deal with a system of $3N$ equations for realistically large $N \rightarrow \infty$, because of which one always

has to invoke some approximation permitting one to reduce the system of untreatably large number of equations to a treatable finite dimensional dynamical system. The most common such a way of reduction is by using the uniform approximation which assumes that the variables u_i and s_i do not depend on the index i . This approximation is known [20,25] to give a good description when there are no electromagnetic spatial structures and the considered sample is sufficiently large for boundary effects being neglected. The uniform approximation reduces equations (18–20) to the same system of equations (25–27). Therefore, the usage of the averages (21), together with the mean-field approximation, is mathematically identical to the uniform approximation. However, from the physical point of view, dealing with the averages is more preferable since an averaging description serves as a reasonable first approximation even when the sample is nonuniform [20,25].

4 Liberation of light

Suppose that at the initial time the atoms are excited so that their average population difference $s_0 \equiv s(0) > 0$. For a single atom, when $g = 0$, from equations (35) one gets $s(t) = s_0$, that is, the emission is suppressed. For a collective of atoms, when $g \neq 0$, the system of equations (35) becomes nonlinear, and its solutions essentially depend on the value of the atom-atom coupling parameter g , defined in equation (24).

According to the notion of suppressed emission, discussed in the Introduction, we need, first, to consider the stationary solutions of the evolution equations. For equations (35), there are two stationary solutions. One of them,

$$s_1^* = s_0, \quad w_1^* = 0, \quad (36)$$

tells that emission is suppressed. And another one,

$$s_2^* = \frac{1}{g}, \quad w_2^* = \frac{\gamma_1(g s_0 - 1)}{4\gamma_2 g^2}, \quad (37)$$

shows that light can, at least partially, be liberated. The asymptotic stability of the stationary solutions (36, 37) can be studied involving the Lyapunov analysis. This is done by calculating the Jacobian matrix associated with equations (35) and finding its eigenvalues that are

$$\lambda^\pm = -\frac{1}{2}[\gamma_1 + \gamma_2(1 - g s)] \mp \frac{1}{2} \left\{ [\gamma_1 - 2\gamma_2(1 - g s)]^2 - 32\gamma_2^2 g^2 w \right\}^{1/2}.$$

These eigenvalues, evaluated at the corresponding fixed points (36, 37), define the characteristic exponents

$$\lambda_1^+ = -\gamma_1, \quad \lambda_1^- = -2\gamma_2(1 - g s_0), \quad \lambda_2^\pm = -\frac{\gamma_1}{2} \left\{ 1 \pm \left[1 + 8\frac{\gamma_2}{\gamma_1}(1 - g s_0) \right]^{1/2} \right\}. \quad (38)$$

The real parts of the expressions in equation (38) are the Lyapunov exponents whose signs characterize the stability of the corresponding fixed points.

The Lyapunov analysis shows that if $g s_0 < 1$, the stationary solution (36) is a stable node, while the fixed point (37) is a saddle point. When $g s_0 = 1$, both fixed points (36, 37), merge together becoming neutral. In the interval

$$1 < g s_0 \leq 1 + \frac{\gamma_1}{8\gamma_2},$$

solution (36) is a saddle point, while the fixed point (37) is a stable node. When

$$g s_0 > 1 + \frac{\gamma_1}{8\gamma_2},$$

the fixed point (36) remains a saddle point, and solution (37) becomes a stable focus with the characteristic exponents

$$\lambda_2^\pm = -\frac{\gamma_1}{2} \mp i\omega_{\text{osc}},$$

where the oscillation frequency is

$$\omega_{\text{osc}} \equiv \frac{\gamma_1}{2} \left[8\frac{\gamma_2}{\gamma_1}(g s_0 - 1) - 1 \right]^{1/2}.$$

This frequency defines the asymptotic oscillation period

$$T_{\text{osc}} \equiv \frac{2\pi}{\omega_{\text{osc}}} = \frac{4\pi}{\sqrt{8(g s_0 - 1)\gamma_1\gamma_2 - \gamma_1^2}}. \quad (39)$$

In the case when $\gamma_1 \ll \gamma_2$, $s_0 \sim 1$, and $g \gg 1$, the latter simplifies to

$$T_{\text{osc}} \simeq 2\pi \sqrt{\frac{T_1 T_2}{2g}},$$

where $\gamma_1 T_1 \equiv 1$ and $\gamma_2 T_2 \equiv 1$, so that T_1 and T_2 are the longitudinal and transverse relaxation times, respectively.

In this way, if the coherent interactions between atoms are weak, and the atom-atom coupling parameter g is small, so that $g s_0 < 1$, then the system tends to the stationary solution (36), that is, there is no liberation of light. When the atomic interactions become stronger, so that $g s_0 > 1$, the system tends to another stable stationary solution (37), and a partial liberation of light occurs since

$$s_2^* = \frac{1}{g} < s_0 \quad (g s_0 > 1).$$

The portion of excitation that remains in the atomic ensemble decreases, with increasing g , as $s_2^* \rightarrow 0$. The qualitative change of the asymptotic behaviour of solutions to equations (35) happens when $g s_0 = 1$. This equality defines the bifurcation point for equations (35), when the dynamical system is structurally unstable [34]. This bifurcation point separates the regions where emission remains suppressed ($g s_0 < 1$) and where light becomes partially liberated ($g s_0 > 1$). For sufficiently strong atomic interactions, such that $g s_0 > 1 + \gamma_1/8\gamma_2$, the liberation of light occurs by means of a series of pulses that, at asymptotically large times, are separated one from another by the period (39).

To describe the coherent pulse, occurring when $\gamma_1 = 0$, we may consider equations (35) omitting there the relaxation term with γ_1 . Then this system of nonlinear equations can be solved exactly resulting in the solution

$$\begin{aligned} s &= -\frac{\gamma_0}{g\gamma_2} \tanh\left(\frac{t-t_0}{\tau_0}\right) + \frac{1}{g}, \\ w &= \frac{\gamma_0^2}{4g^2\gamma_2^2} \operatorname{sech}^2\left(\frac{t-t_0}{\tau_0}\right), \end{aligned} \quad (40)$$

where the radiation width γ_0 is given by the relation

$$\gamma_0^2 = \Gamma_0^2 + 4g^2\gamma_2^2 (|u_0|^2 - \alpha_0 s_0^2) \quad (41)$$

in which $u_0 \equiv u(0)$, $\alpha_0 \equiv \alpha(0)$, and

$$\Gamma_0 \equiv \gamma_2(1 - gs_0), \quad \gamma_0 \equiv \frac{1}{\tau_0},$$

and where the delay time is

$$t_0 = \frac{\tau_0}{2} \ln \left| \frac{\gamma_0 - \Gamma_0}{\gamma_0 + \Gamma_0} \right|. \quad (42)$$

Introducing the critical atom-polariton coupling

$$\alpha_c \equiv \frac{(gs_0 - 1)^2}{4g^2s_0^2} + \frac{|u_0|^2}{s_0^2}, \quad (43)$$

the radiation width can be written as

$$\gamma_0 = 2g|s_0|\gamma_2\sqrt{\alpha_c - \alpha_0}. \quad (44)$$

The value (43) is termed critical since the coupling α_0 cannot exceed α_c for the solution (40) to remain finite. The restriction $\alpha_0 \leq \alpha_c$ specifies condition (31) assumed for the validity of the averaging method. This restriction is not as severe as far as $\alpha \ll 1$ while, for large g , $\alpha_c \geq 1/4$.

At the time $t = t_0$, the solutions (40, 34) yield

$$\begin{aligned} s(t_0) &= \frac{1}{g}, \quad w(t_0) = s_0^2(\alpha_c - \alpha_0), \\ \langle\langle |u(t_0)|^2 \rangle\rangle &= s_0^2(\alpha_c - \alpha_0) + \frac{\alpha}{g^2}. \end{aligned} \quad (45)$$

Then the system of atoms achieves the maximum of coherence.

Consider more in detail the case when the system of atoms is initially completely inverted, $s_0 = 1$, there is no triggering pulse, $u_0 = 0$, and the atom-atom coupling is strong, $g > 1$. Then the critical value (43) becomes

$$\alpha_c = \left(\frac{g-1}{2g}\right)^2. \quad (46)$$

If $\alpha \ll \alpha_c$, the radiation width (44) gives

$$\gamma_0 = (g-1)\gamma_2 \left[1 - \frac{2g^2\alpha_0}{(g-1)^2} \right]. \quad (47)$$

The corresponding radiation time is

$$\tau_0 = \frac{T_2}{g-1} \left[1 + \frac{2g^2\alpha_0}{(g-1)^2} \right]. \quad (48)$$

And for the delay time (42), we have

$$t_0 = \frac{T_2}{2(g-1)} \ln \left| \frac{(g-1)^2 - g^2\alpha_0}{g^2\alpha_0} \right|. \quad (49)$$

When the atomic coupling is strong, then

$$\alpha_c \simeq \frac{1}{4} \quad (u_0 = 0, g \gg 1). \quad (50)$$

The radiation time (48) for $g \gg 1$ becomes

$$\tau_0 \simeq \frac{T_2}{g} (1 + 2\alpha_0). \quad (51)$$

Hence, τ_0 can be much smaller than T_2 . Since $g \sim N$, we have $\tau_0 \sim N^{-1}$, which is typical of superradiance [20,25]. Under condition (50), the delay time (49) is

$$t_0 \simeq \frac{T_2}{2g} |\ln \alpha_0|. \quad (52)$$

Note that if $\alpha_0 \rightarrow 0$, then $t_0 \rightarrow \infty$, which means that emission would be suppressed for very long time. The superradiant burst develops at the delay time t_0 and lasts during the radiation time τ_0 .

In the case when $\alpha_c - \alpha_0 \ll 1$, the radiation time increases, as compared to the opposite case when $\alpha_0 \ll \alpha_c$, being

$$\tau_0 = \frac{T_2}{2g|s_0|\sqrt{\alpha_c - \alpha_0}}, \quad (53)$$

while the delay time shortens becoming

$$t_0 = \frac{2g|s_0|T_2}{(gs_0 - 1)^2} \sqrt{\alpha_c - \alpha_0}. \quad (54)$$

In this case, the radiation can be coherent if $\tau_0 \ll T_2$, which requires $g\sqrt{\alpha_c - \alpha_0} \gg 1$.

The superradiant character of emission is connected with the radiation time being inversely proportional to the number of radiators, $\tau_0 \sim N^{-1}$. Another characteristic describing the level of coherence in the radiating system is the radiation intensity. The total intensity of radiation, averaged over fast oscillations,

$$I(t) = I_{\text{inc}}(t) + I_{\text{coh}}(t), \quad (55)$$

consists of two terms, the intensity of incoherent radiation

$$I_{\text{inc}} = \frac{1}{2}\omega_0\gamma N(1+s) \quad (56)$$

and the intensity of coherent radiation

$$I_{\text{coh}} = \omega_0\gamma\varphi_s N^2 \langle\langle |u|^2 \rangle\rangle, \quad (57)$$

where φ_s is the shape factor [35] given by the integral over spherical angles,

$$\varphi_s = \frac{3}{8\pi} \int |\mathbf{n} \times \mathbf{e}_d|^2 F(k_0 \mathbf{n}) d\Omega(\mathbf{n}),$$

$$F(\mathbf{k}) \equiv \left| \frac{1}{N} \sum_{i=1}^N e^{i\mathbf{k} \cdot \mathbf{r}_i} \right|^2. \quad (58)$$

For the point-like system, for which $k_0 \rightarrow 0$, we have $\varphi_s \rightarrow 1$, and the intensity of coherent radiation is proportional to the number of radiators squared, $I_{\text{coh}} \sim N^2$, which is typical of the Dicke model. For a finite-size system, the shape factor essentially depends on the relation between the radiation wavelength λ and the characteristic sizes of the sample [35]. Thus, for a cylindrical sample of radius R and length L , we have

$$\varphi_s \simeq \begin{cases} \frac{3\lambda}{8L}, & \frac{\lambda}{2\pi L} \ll 1, \frac{R}{L} \ll 1, \\ \frac{3}{8} \left(\frac{\lambda}{\pi R} \right)^2, & \frac{\lambda}{2\pi R} \ll 1, \frac{L}{R} \ll 1 \end{cases}$$

for pencil-like or disk-like shapes, respectively. Hence for the first case $I_{\text{coh}} \sim N^{5/3}$, while for the second case $I_{\text{coh}} \sim N^{4/3}$.

To compare the intensities of coherent and incoherent radiation, it is convenient to introduce the coherence coefficient [36] defined as the ratio

$$C_{\text{coh}}(t) \equiv \frac{I_{\text{coh}}(t)}{I_{\text{inc}}(t)}. \quad (59)$$

For the coherent radiation intensity (57), using the relation (34), we get

$$I_{\text{coh}}(t) = \omega_0 \gamma \varphi_s N^2 (w + \alpha s^2).$$

Therefore, the coherence coefficient (59) is

$$C_{\text{coh}} = 2\varphi_s N \frac{w + \alpha s^2}{1 + s}. \quad (60)$$

At the moment of the maximal coherence of the superradiant burst, according to equations (45), we find

$$C_{\text{coh}}(t_0) = 2g\varphi_s s_0^2 N \frac{\alpha_c - \alpha_0}{1 + g}.$$

If $C_{\text{coh}} > 1$, the radiation is predominantly coherent. For the case when $u_0 = 0$, $s_0 = 1$, and $g \gg 1$, the latter expression gives

$$C_{\text{coh}}(t_0) \simeq \frac{1}{2} \varphi_s N.$$

If the sample has a pencil-like or disk-like shape, then

$$C_{\text{coh}}(t_0) \simeq \begin{cases} \frac{3\pi}{16} \left(\frac{R}{\lambda} \right)^2 \rho \lambda^3, & \text{pencil} \\ \frac{3}{16\pi} \left(\frac{L}{\lambda} \right) \rho \lambda^3, & \text{disk,} \end{cases}$$

where ρ is the density of resonance atoms. When this density is sufficiently high, and $\lambda \ll R$ or $\lambda \ll L$, the coher-

ence coefficient can be very large, which would mean that the radiation is almost purely coherent.

The general condition for light to be, at least partially, liberated is

$$s(\infty) < s_0. \quad (61)$$

From equations (40), we have

$$s(\infty) = -\frac{\gamma_0}{g\gamma_2} + \frac{1}{g}, \quad w(\infty) = 0,$$

which, with the radiation width (44), gives

$$s(\infty) = -2|s_0| \sqrt{\alpha_c - \alpha_0} + \frac{1}{g}. \quad (62)$$

Therefore the liberation condition (61) becomes

$$g(s_0 + 2|s_0| \sqrt{\alpha_c - \alpha_0}) > 1. \quad (63)$$

In the case, when $u_0 = 0$, and taking into account that $\alpha_0 \ll 1$, equation (62) reduces to

$$s(\infty) = \begin{cases} s_0, & gs_0 < 1 \\ -s_0 + 2/g, & gs_0 > 1. \end{cases} \quad (64)$$

The condition (63) simplifies to

$$gs_0 > 1. \quad (65)$$

It is interesting that, although the limit values $s(\infty)$ are different for the cases when γ_1 is finite or when it is zero, but the liberation condition (65) remains the same. Under this condition, a system of atoms can radiate, though spontaneous emission of a single atom is suppressed. The case of a single atom can be recovered by setting $g \rightarrow 0$. Then, as is obvious, condition (65) can never hold true. It is only when the density of doped atoms is sufficiently high, so that the atom-atom coupling g becomes sufficiently large, satisfying condition (65), the radiation of atoms is possible being due to collective effects.

5 Discussion

We considered a system of resonance atoms doped into a medium with a polariton band gap. Spontaneous emission of a single atom with the transition frequency inside the polariton gap is suppressed. However an ensemble of atoms with their transition frequencies in the gap can radiate due to effective coherent interactions between the atoms. If this interaction is sufficiently strong, light is partially liberated. The collective liberation of light occurs through one or a series of superradiant pulses.

The dynamics of light liberation has been analysed employing the scale separation approach [30–32], which is a generalization of the averaging method [33] to stochastic differential equations. This approach provides an efficient tool for treating complicated systems of nonlinear evolution equations, as has earlier been demonstrated for the

problems of superradiant spin relaxation in nonequilibrium magnets [30–32] and of nonlinear dynamics of atoms in magnetic traps [37–39], where nonlinear phenomena are of crucial importance. Collective liberation of light is also a principally nonlinear phenomenon, with nonlinearity caused by coherent atomic interactions.

In conclusion, let us give some estimates for the characteristic parameters considered in the text. The polariton effect is well developed in many dielectrics and semiconductors [4,5]. For instance, it is intensively studied in such semiconductors as CuCl, CuBr, CdSe, ZnSe, GaAs, GaSb, InAs, AlAs, and SiC [10–12,40]. The polariton gap in such materials develops around the frequency 10^{14} s^{-1} , with the gap width $\Delta_p \sim 10^{13} \text{ s}^{-1}$. By assumption, the atomic transition frequency is inside the polariton gap, *i.e.* $\omega_0 \sim 10^{14} \text{ s}^{-1}$. Hence the radiation wavelength is $\lambda \sim 10^{-3} \text{ cm}$. For the line width, one may take $\gamma_2 \sim 10^9 \text{ s}^{-1}$.

For the initially inverted atoms, with $s_0 = 1$, the critical atom-atom coupling parameter, above which the collective radiation becomes possible, is $g_c = 1$. For the parameters $g > g_c$, the collective liberation of light occurs. If $\gamma \sim \gamma_2$, then the atomic coupling parameters, defined in equation (24), are $g \sim \rho\lambda^3$ and $g' \sim \rho\lambda^3$, where ρ is the density of atoms. Consequently, the critical density of atoms, providing $g_c = 1$, is $\rho_c \sim \lambda^{-3}$, which gives $\rho_c \sim 10^9 \text{ cm}^{-3}$. When $\rho > \rho_c$, radiation becomes possible because of the formation of an impurity band inside the polariton band. The width of the impurity band is of the order of the collective line width $\Gamma \sim \gamma_2 g \sim \gamma\rho\lambda^3$. The latter becomes larger than the polariton gap, when the density $\rho > \Delta_p/\gamma_2\lambda^3$, that is, $\rho > 10^{13} \text{ cm}^{-3}$. For such densities of resonance atoms, the polariton gap can be overlapped by the impurity band. At the density $\rho \sim 10^{13} \text{ cm}^{-3}$, the atom-atom coupling is $g \sim 10^4$, and the collective line width is $\Gamma \sim 10^{13} \text{ s}^{-1}$.

With the densities $\rho < 10^{13} \text{ cm}^{-3}$, the atom-atom coupling $g < 10^4$, while the atom-polariton coupling is $\alpha \sim \gamma^2/\Delta_p^2$, that is $\alpha \sim 10^{-8}$. The radiation time $\tau_0 \sim T_2/g$ is larger than 10^{-13} s but can be much smaller than $T_2 \sim 10^{-9} \text{ s}^{-1}$. And the delay time $t_0 \sim \tau_0 |\ln \alpha|$ is an order longer than the radiation time. In this way, for the density of doped atoms $\rho \sim 10^9\text{--}10^{13} \text{ cm}^{-3}$, the effective atom-atom coupling is $g \sim 1\text{--}10^4$. Then the delay time is $t_0 \sim 10^{-12}\text{--}10^{-8} \text{ s}$ and the radiation time $\tau_0 \sim 10^{-13}\text{--}10^{-9} \text{ s}$.

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