Superradiance and Subradiance in an Inhomogeneously Broadened Ensemble of Two-Level Systems Coupled to a Low-Q Cavity

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The collective spontaneous emission of a fully inverted inhomogeneously broadened ensemble of N two-level systems coupled to a single-mode low-Q cavity is investigated numerically using Monte Carlo wave function technique. An intrinsically bi-exponential emission dynamics is found when the time scales of superradiance τ_{sr} and inhomogeneous dephasing $T_2^* \sim 1/\Delta\omega_{inh}$ become comparable: a fast superradiant is followed by a slow subradiant decay. Experimental configurations using ensembles of quantum dots coupled to optical microcavities are proposed as possible candidates to observe the combined superradiant and subradiant energy relaxation.

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The collective spontaneous emission of an ensemble of N excited two-level systems predicted by Dicke in 1954 [1] represents one of the most complicated and extensively investigated fundamental effects in quantum optics [2–4]. If the relaxation time of the polarization T_2 is long enough, the energy stored in the atomic system can be released into the field mode via *collective* spontaneous emission at a certain rate, which depends on the initial state of the system. For example, if all atoms are initially inverted, the first spontaneously emitted photons trigger a buildup of a large macroscopic atomic polarization during the emission of a superfluorescent pulse. The peak intensity and emission rate of such a superfluorescent pulse is $\sim N$ times larger compared to that of N independent atoms [1], which in a classical picture corresponds to parallel dipole moments of all radiating atoms [4]. However, there also exist the so-called subradiant states, in which the energy remains partially trapped in the atomic system since some of the atomic dipoles become antiparallel and the macroscopic polarization of the system is zero; i.e., they cannot couple to a radiation field [1].

If the two-level systems are *identical* the permutational symmetry of the atom-field interaction Hamiltonian rules out the coupling between the superradiant and subradiant states. However, any type of symmetry breaking allows the coexistence of superradiance and subradiance in the sense that the system initially prepared in a superradiant state will partially evolve into the subradiant state. Such behavior is particularly important for quantum optics of zero-dimensional semiconductor nanostructures (e.g., quantum dots, often referred to as ''artificial atoms'') [5], which are inevitably *different* in their transition frequencies and oscillator strengths due to the peculiarities of the manufacturing process [6]. Very recently, the generalized Dicke model including the dipole-dipole coupling attracted a renewed interest motivated by the possibility to observe superradiance and subradiance in an ensemble of very densely spaced quantum dots [7].

In this Letter we investigate numerically the dynamics of an inhomogeneously broadened, fully inverted ensemble of N two-level systems coupled to a short-living cavity mode. We predict that a strong inhomogeneous broadening in the Dicke model leads to an intrinsically bi-exponential temporal evolution of the emission dynamics in which a fast superradiant decay is followed by a slow subradiant energy relaxation. We discuss the implications of this result for time-resolved emission experiments on strongly confined quantum dots coupled to optical microcavities with low quality factor Q, like those sketched in Fig. 1.

The effect of inhomogeneous broadening upon superradiance was investigated in atomic physics in the 1970s both theoretically and experimentally [2–4]. Some theoretical results were obtained by Eberly [8,9], Agarwal [10],

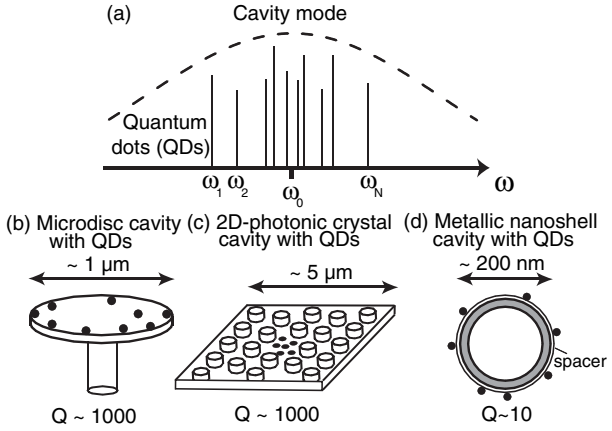

FIG. 1. An inhomogeneously broadened ensemble of N two-level systems coupled to a single damped cavity mode: general schematic in frequency domain (a) and possible experimental configurations in semiconductor cavity QED such as semiconductor quantum dots in a microdisc cavity (b) or 2D-photonic crystal cavity (c), or semiconductor nanocrystals linked via spacer molecules to the surface of a metallic nanoshell cavity (d).

Bonifacio and Lugiato [11] within the semiclassical approximation. Leonardi and Vaglica [12] developed a fully quantum mechanical theory of superfluorescence for a weak inhomogeneous broadening, i.e.,

$$T_2^* \gg \sqrt{\tau_{sr}\tau_d}, \tag{1}$$

holds, where T_2^* is the inhomogeneous dephasing time and $\tau_{\rm sr}$ and τ_d are the duration and delay time of superfluorescent pulses. According to [12] most of the experiments carried out at that time satisfied the condition (1) and thus could be adequately interpreted using the semiclassical approximation as, for example, was demonstrated by Haake *et al.* [13]. In this work we investigate the case of strongly inhomogeneous dephasing, when the condition (1) is not satisfied.

The general scheme of the model system in the frequency domain is sketched in Fig. 1(a). The collection of N two-level systems (to be referred to as atoms) with different resonance frequencies ω_n is coupled to a single, strongly damped cavity mode with a short photon lifetime $(2\kappa)^{-1}$ centered at frequency ω_0. The homogeneous linewidth of individual atomic transitions in free space is assumed to be small and is neglected in the simulations ($T_2' \to \infty$). The atomic detunings $\delta\omega_n = \omega_n - \omega_0$ obey a Gaussian distribution with the width (FWHM) $\Delta\omega_{\rm inh} \sim 1/T_2^*$. The interaction between the atoms and the field is described by the Tavis-Cummings Hamiltonian [14]:

$$H_{\rm AF} = i\hbar g[aJ_+(t) - a^\dagger J_-(t)], \tag{2}$$

where the time-dependent operators for collective atomic polarization $J_\pm(t) = \sum_{n=1}^N \sigma_\pm^{(n)} \exp(\pm i\delta\omega_n t)$ explicitly incorporate the effect of inhomogeneous broadening [11]; g is the atom-field coupling constant, a and $a^\dagger$ are the annihilation and creation operators of photons in the cavity mode, $\sigma_\pm^{(n)} = \sigma_x^{(n)} \pm i\sigma_y^{(n)}$, with $\sigma_{x,y,z}^{(n)}$ representing Pauli matrices for individual atoms. The following master equation for the atom-field density operator $\rho_{\rm AF}$,

$$\frac{d\rho_{\rm AF}}{dt} = -\frac{i}{\hbar}[H_{\rm AF}, \rho_{\rm AF}] + \kappa(2a\rho_{\rm AF}a^\dagger - a^\dagger a\rho_{\rm AF} - \rho_{\rm AF}aa^\dagger) \tag{3}$$

with κ being the damping constant of the cavity mode, describes the temporal evolution of the system. In a bad-cavity limit

$$\frac{g^2 N}{\kappa}, \qquad \Delta\omega_{\rm inh} \ll \kappa \tag{4}$$

the cavity mode can be eliminated adiabatically and (3) can be reduced to the master equation of superradiance for the atomic density operator $\rho = {\rm tr}_F \rho_{\rm AF}$ [10]

$$\frac{d\rho}{dt} = \frac{g^2}{\kappa}[2J_-(t)\rho J_+(t) - J_+(t)J_-(t)\rho - \rho J_-(t)J_+(t)]. \tag{5}$$

In the overdamped limit the intensity of radiation $I(t)$ emitted into cavity mode is proportional to the squared atomic polarization:

$$I(t) = I_1 \hbar\omega_0 \langle J_+(t)J_-(t)\rangle, \tag{6}$$

where $I_1 = 2g^2/\kappa$ stands for a single-atom rate of spontaneous emission in the cavity mode and a commonly used assumption $\Delta\omega_{\rm inh} \ll \omega_0$ is exploited [10]. The squared atomic polarization consists of two terms

$$\langle J_+(t)J_-(t)\rangle = \sum_{n=1}^N \langle \sigma_+^{(n)}\sigma_-^{(n)}\rangle(t) + \sum_{n\neq m}^N \langle \sigma_+^{(n)}\sigma_-^{(m)}\rangle(t) e^{i(\omega_n - \omega_m)t}, \tag{7}$$

where the first term represents the sum of independent emission rates of atoms while the second displays pair correlations between atomic dipoles. Positive correlations (parallel dipoles) are characteristic for superradiance whereas negative ones (antiparallel dipoles) display subradiance. For this reason the second term in Eq. (7), denoted as $\langle J_+J_-\rangle_{\rm corr}$, deserves special attention. In most previous reports [8–11] the pair correlations $\langle \sigma_+^{(n)}\sigma_-^{(m)}\rangle$ were assumed to be equal for all pairs of atoms, which allows the semiclassical analytical solutions to be obtained but restricts the possible solutions to the symmetric atomic states only [12]. In this work this crucial assumption is avoided and exact numerical solutions of the master Eq. (5) are analyzed.

Temporal evolution of the system can be also visualized following the decay of the total energy

$$W(t) = \sum_{n=1}^N \left[\langle \sigma_z^{(n)}\rangle(t) + \frac{1}{2}\right] = \sum_{k=0}^N k p_k(t), \tag{8}$$

which obeys the energy conservation law $\hbar\omega_0 {\rm d}W/{\rm d}t = I(t)$ and is equal to the expectation value of the total atomic inversion shifted by $N/2$; $p_k(t)$ represent the probabilities of the atomic configuration with k inverted atoms.

The master equation (5) was solved numerically using the Monte Carlo wave function technique [15], which implies the propagation in time of a stochastic wave function with 2^N complex components [16]. The simulations were performed for a reasonably large atom number $N = 10$. The inhomogeneous broadening was simulated by randomly distributing of 10 atomic transition frequencies ω_n according to a Gaussian distribution. The results obtained for different random realizations of ω_n are qualitatively similar and only slightly different for $N = 10$. However, in order to rule out possible minor effects due to a specific ensemble of frequencies, all quantities are averaged over many random realizations of frequency distributions. At zero time all atoms are inverted and temporal evolution of the system is investigated as a function of the parameter

$$\alpha = \frac{I_1 N}{2\Delta\omega_{\rm inh}} = \frac{T_2^*}{\tau_{\rm sr}}, \tag{9}$$

which represents the ratio of the inhomogeneous dephasing time T_2^* to the typical time scale of superradiance $\tau_{\rm sr}$. The

condition (1) is equivalent to $\alpha \gg 1$ since the difference between the pulse duration $\tau_{sr} = 2I_1/N$ and the moment in time $\tau_d = 0.5\tau_{sr} \ln N$ when the maximum is reached [both times scales appear in Eq. (1)] becomes significant only for a very large N.

The temporal evolution of the squared atomic polarization $\langle J_+ J_- \rangle$ is shown in Fig. 2(a) for different values of α. In the case of negligible inhomogeneous broadening $T_2^* \gg \tau_{sr}$ ($\alpha = 10$), a superfluorescent pulse is emitted: the polarization starts growing, reaches the maximum at $t \approx \tau_{sr}$, and rapidly decays to zero. As α decreases the amplitude of the pulse becomes smaller and the behavior converges to the second limiting case of uncorrelated spontaneous emission $\sim \exp(-I_1 t)$ for $\alpha \ll 1$. When inspecting the dynamics of $\langle J_+ J_- \rangle$ on a logarithmic scale in Fig. 2(b) a remarkable bi-exponential behavior becomes evident for $\alpha \sim 1$.

More detailed information can be inferred from Fig. 3, where the dynamics of atomic energy W are shown on a logarithmic scale together with the correlated part of squared atomic polarization $\langle J_+ J_- \rangle_{corr}$. In case of negligible inhomogeneous dephasing, $\alpha = 10$, the atomic energy rapidly decays to zero with a superradiant rate $\sim I_1 N$. The correlated part $\langle J_+ J_- \rangle_{corr}$ of superradiant spontaneous emission remains positive for all times, whereas for the uncorrelated case $\alpha = 0$ it is zero by definition. A bi-exponential energy decay is again observed for $\alpha \sim 1$: the first fast superradiant component for $t < 5\tau_{sr}$ is followed by a second slow component for $t > 5\tau_{sr}$. Positive pair correlations $\langle J_+ J_- \rangle_{corr}$ build up in the beginning of the emission process, which explains a fast initial superradiant decay rate. For $\alpha \sim 1$, $\langle J_+ J_- \rangle_{corr}$ becomes negative at a time $t \simeq 5\tau_{sr}$, at which the energy decay rate changes from fast to slow. The change in sign of $\langle J_+ J_- \rangle_{corr}$ corresponds to the antiparallel orientation of some atomic dipoles and favors the subradiant behavior. For longer delay times the negative correlations decay to zero when antiparallel dipoles of individual atoms having slightly different frequencies run out of phase. The second slow energy relaxation rate in Fig. 3 is always significantly smaller than I_1 for $\alpha \sim 1$. It monotonously decreases when N is increased or T_2^* decreased. In order to further clarify the physical nature of the slow subradiant relaxation we follow the temporal evolution of the probabilities $p_k(t)$ for $\alpha = 1$ in Fig. 4. All $p_k(t)$ grow, starting from zero [except for $p_{10}(t)$; $p_{10}(0) = 1$], on a fast superradiant time scale and decay on a time scale which strongly depends on the number of inverted atoms k. The population $p_1(t)$ of collective states with only one inverted atom decays exponentially with the longest time constant.

Monte Carlo simulations of the more general master equation (3) show that bi-exponential behavior of superradiance and subradiance persists even if the condition (4) is not fulfilled (i.e., $I_1 N$, $\Delta\omega_{inh} \sim \kappa$). This extends the applicability of our simulations to an interesting case where the cavity mode only interacts with a subensemble of the emitters that falls within its spectral width.

Several realistic experimental configurations, which are sketched in Fig. 1, are proposed as possible candidates to investigate the collective spontaneous emission from a

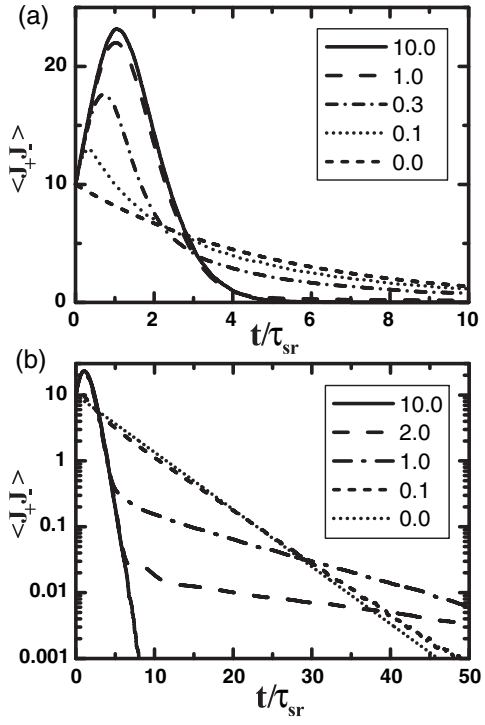

FIG. 2. Temporal evolution of squared polarization $\langle J_+ J_- \rangle$ for an ensemble of 10 initially inverted atoms for different α on a linear scale (a) and a logarithmic scale (b) ($\alpha = 10$ corresponds to superradiance; $\alpha = 0$ represents uncorrelated spontaneous emission).

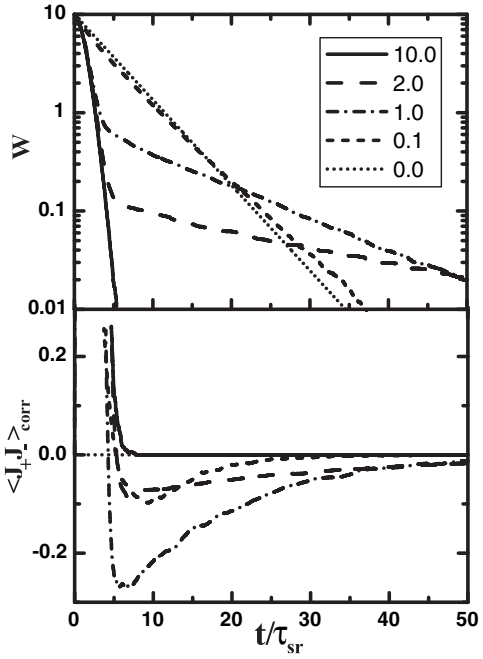

FIG. 3. Temporal evolution of atomic energy W and correlated part of atomic polarization $\langle J_+ J_- \rangle_{corr}$ for different α.

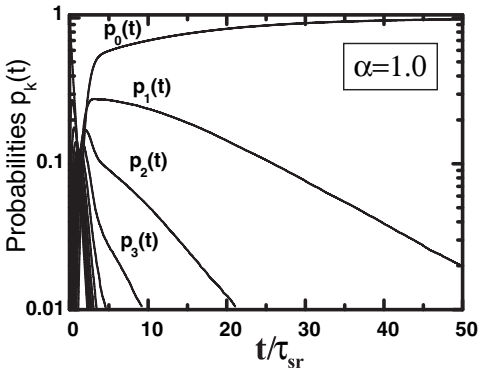

FIG. 4. Dynamics of the probabilities $p_k(t)$ of having k inverted atoms for $\alpha = 1$ (at $t = 0$ all 10 atoms are inverted).

quantum dot ensemble coupled to a damped single-mode optical cavity. At helium temperatures the intrinsic dephasing of strongly confined quantum dot two-level systems due to electron-phonon coupling can be suppressed and the homogeneous lifetime T_2' is limited only by the lifetime of inversion T_1, which is of the order of 1 ns for single quantum dots in free space [17]. The coupling to a cavity under the condition of the strong Purcell effect makes it possible to direct the spontaneous emission of two-level-like quantum dots predominantly into the single cavity mode and manipulate its rate. For microcavities shown in Fig. 1 the quantum dot density can be chosen to be sufficiently low to avoid direct dipole-dipole interaction via the near field of oscillating dipoles. Moreover, the dipole-dipole coupling, which is neglected in Dicke model, is argued to be less important for superradiance in a cavity [18].

Microdisc and 2D-photonic crystal cavities [Fig. 1(b) and 1(c)] can be manufactured with small mode volumes $V_m \sim \lambda^3$ and relatively low $Q \sim 1000$, which favors the condition of the strong Purcell effect (Purcell factor $F_p \gg 1$) [19]. In such cavities the single-dot spontaneous emission time into a cavity mode can be reduced to several tens of picoseconds. According to our calculations the superradiant emission would then occur on time scale of a few picoseconds whereas the subradiant emission would last some hundreds of picoseconds. When quantum dots are excited nonresonantly by an ultrashort laser pulse and the emission dynamics then measured with picosecond time resolution at low temperature, it should be possible to observe both superradiant and subradiant decay rates from the cavity, which are significantly above or below the single-dot emission rate.

A different type of a plasmonic nanocavity in Fig. 1(d) consists of a dielectric core covered by a thin metallic shell and provides broad tunable plasmon modes with small $Q \sim 10$ [20]. Despite the much smaller Q factor, the spontaneous emission into the plasmon cavity mode is also likely to dominate over the one in free space, since the latter may be strongly suppressed in the presence of a metallic surface [21]. Thus such type of cavity may be suitable to investigate cooperative spontaneous emission in a plasmon mode.

In conclusion, the cooperative spontaneous emission of a fully inverted ensemble of N two-level systems in a single damped cavity mode in the presence of strong inhomogeneous broadening shows combined superradiant and subradiant emission dynamics. Semiconductor quantum dot ensembles coupled to a single-mode optical cavity are proposed as model systems to demonstrate experimentally these fundamental effects in cavity QED.

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