Laser Control of Collective Spontaneous Emission

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The collective spontaneous emission of a pair of two coupled three-level radiators in vacuum is investigated in the presence of a possibly intense laser field. The parameters describing the collective interaction along with the population and decay rates of all involved dressed states are shown to be controllable by the applied laser field. In particular, all populations of the collective system may be transferred at will in a reversible way into a subradiant state, allowing effective storage and manipulation of the quantum system.

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Spontaneous emission is one of the most frequently occurring physical processes and as such its control is of vital importance also for almost every stimulated operation. In the case of single atoms there is considerable experience in inhibiting the mostly unwanted influence of the environment. A very successful way to modify the interacting part of the environment has proven to be effective via changing the involved atomic transition frequencies [1] or directly placing the atom into a cavity [2], an optical waveguide [3], a photonic band gap material [4], or an otherwise modified vacuum [5]. The decay of an excited state may equally be reduced by repeated measurements, i.e., the quantum Zeno effect [6], or by destructive quantum interference, mostly via additional laser fields [7]. Here spontaneous emission was shown to be virtually completely inhibited at will or arbitrarily narrow structures could be shaped with variable intensity.

For collective atomic systems, the possibility of superradiance or subradiance, i.e., substantial enhancements and reductions of the total spontaneous emission rates are also known for quite some time [8–11]. However, especially nondecaying ensembles of atoms are difficult to prepare because they are decoupled from the environment in general. The interest in those systems is considerable though, because entangled states involving at least two two-state systems play a vital role in quantum information theory [12,13] such as in atomic traps [14]. Decay and other means of incoherently induced decoherence are critical as they generally mean a loss of information.

In this Letter a method is proposed which is suitable to manipulate the collective spontaneous emission of a pair of two three-level radiators with the help of a possibly strong laser field. The various effective semiclassical dressed atomic states are determined via the laser-atom and atom-atom couplings (see Fig. 1) and their Schrödinger dynamics is evaluated with regard to their steady-state populations and spectral properties. The strong interaction of the laser field with the atoms is shown to modify the atom-vacuum interaction in case of single-atom resonance fluorescence, and especially indirectly the collective parameters describing the mutual interaction between the atoms. This arises due to the fact that the controllable dressed atomic transitions may interact with different parts of the vacuum and consequently also affect the mutual dipole-dipole coupling of the atoms. Finally, with the additional inclusion of incoherent pumping, emphasis is placed on how to transfer the population completely into an antisymmetrical nondecaying two-atom collective state and thus to prevent the system from decaying.

Our system is characterized by two identical nonoverlapping three-level "ladder" radiators (A and B) at positions $\vec{r}_A$ and $\vec{r}_B$ and separation $\vec{r}_{AB}$ in interaction with an external resonant coherent field (see Fig. 1). The dipoles of both atoms are assumed parallel and fixed, e.g., via a surrounding organic layer or a cavity [15]. The atomic transition between the states $|3\rangle$ and $|2\rangle$ is driven by a coherent field of wave vector $\vec{k}_l$ and frequency ω_l, while the other dipole-allowed transition $|2\rangle \rightarrow |1\rangle$ is merely coupled by vacuum modes with a dipole moment d_{21}. In the dipole approximation the Hamiltonian that describes

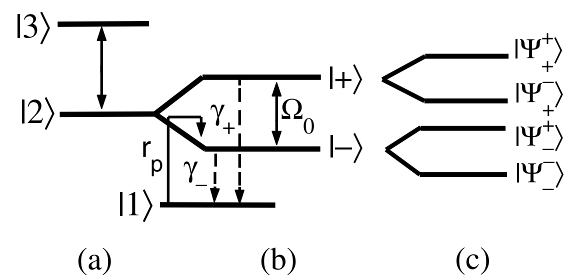

FIG. 1. Schematic configuration depicting (a) the bare-state atomic three-level system, (b) its dressed-state representation via the driving laser field with Rabi frequency Ω_0 and decay rates γ_+ and γ_-, and (c) the two-atom collective dressed states. Initially, atom A is in its bare state $|2\rangle_A$ while the atom B is in the bare state $|1\rangle_B$, and the incoherent pumping r_p is initially set equal to zero. The population from the states $|\Psi_\pm^+\rangle$ and $|\Psi_\pm^-\rangle$ may decay to the state $|\Psi_g\rangle = |\tilde{1}_A, \tilde{1}_B\rangle$.

such an atomic medium can be represented as follows:

$$H = \sum_k \hbar\omega_k a_k^\dagger a_k + \sum_{j\in\{A,B\}}\sum_{\alpha=1}^{3}\hbar\omega_\alpha R_{\alpha\alpha}^{(j)} + \frac{1}{2}\hbar\Omega_0 \sum_{j\in\{A,B\}}\{R_{32}^{(j)} e^{i(\vec{k}_l\cdot\vec{r}_j)-i\omega_l t} + \text{H.c.}\} + i\sum_k \sum_{j\in\{A,B\}}(\vec{g}_k\cdot\vec{d}_{21})\{a_k^\dagger R_{12}^{(j)} e^{-i(\vec{k}\cdot\vec{r}_j)} - \text{H.c.}\}, \quad (1)$$

where $\hbar\omega_\alpha$ are the energies of levels α for $\alpha \in \{1, 2, 3\}$, $a_k^\dagger$ (a_k) are the creation (annihilation) operator for photons with momentum $\hbar\vec{k}$ and energy $\hbar\omega_k$, while $\vec{g}_k$ describe the vacuum-atom coupling (decay of $|3\rangle$ assumed negligible). $R_{\alpha\beta}^{(j)} \equiv |\alpha\rangle_j\langle\beta|$ are the corresponding operators of the transitions among the states $|\beta\rangle$ and $|\alpha\rangle$ of the jth radiator, and satisfy the commutation relation $[R_{\alpha\beta}^{(j)}, R_{\beta'\alpha'}^{(m)}] = \delta_{jm}[\delta_{\beta\beta'}R_{\alpha\alpha'}^{(j)} - \delta_{\alpha\alpha'}R_{\beta'\beta}^{(j)}]$ for $\{\alpha,\alpha',\beta,\beta'\}\in\{1,2,3\}$ and $\{j, m\} \in \{A, B\}$.

In Eq. (1) the first and the second terms represent the free electromagnetic fields (EMF) and the unperturbed atomic eigenstates while the third and fourth expressions describe the interaction of the atomic sample with the laser field and the environmental EMF modes, respectively. Single-atom radiative shifts are negligible here, and we confine our attention to driving laser field intensities with Rabi frequencies larger than the relaxation parameters. As a consequence, it is advantageous to employ laser-dressed states [16] $|\pm\rangle_{A,B} = \frac{1}{\sqrt{2}}\{|2\rangle_{A,B} \pm |3\rangle_{A,B}\}$ and $|\tilde{1}\rangle_{A,B} = |1\rangle_{A,B}$ rather than the bare states $|2\rangle$ and $|3\rangle$. Dipole-dipole coupling is implicitly included in the Hamiltonian in Eq. (1) and appears explicitly in the dressed-state equations of motion. For simplicity we further assume $(\vec{k}_l \cdot \vec{r}_{AB}) = 0$.

We concentrate on the calculation of the collective spontaneous emission spectrum on the $|2\rangle \to |1\rangle$ transition when initially the first atom A is in its bare state $|2\rangle_A$ while the other atom B is in the bare state $|1\rangle_B$ (initially without incoherent pumping). At the point of observation $\vec{r}$, the spontaneously generated spectrum S as a function of the frequency ω can be obtained by evaluating the real part of the Fourier transform of the correlation function of the EMF

$$S(\omega) \propto \int_0^\infty d\tau e^{i\omega\tau}\lim_{t\to\infty}\langle E^{(-)}(\vec{r}, t)E^{(+)}(\vec{r}, t-\tau)\rangle, \quad (2)$$

where $E^{(-)}$ and $E^{(+)}$ represent the positive and negative parts of the EMF operator $\vec{E}$ [17]. Applying the dressed-state representation in Eq. (1) one can diagonalize the unperturbed part of the Hamiltonian [18]. In representing the EMF correlation function in Eq. (2) by operators in the dressed states basis we may neglect terms which oscillate with frequencies higher than the Rabi frequency. Solving the equations of motion for the dressed atomic operators also in the secular approximation and applying the regression quantum theorem [17], we obtain the following expression for the emitted spectrum:

$$S(\tilde{\omega}) = \frac{1}{8}\Phi(\vec{r})\sum_{\mp}\gamma_{\mp}^2\left[\frac{(1+\cos\Theta_{\mp})(1+\chi_{AB}^{\mp})}{[\frac{1}{2}\gamma_{\mp}(1+\chi_{AB}^{\mp})]^2 + [\tilde{\omega} \pm \frac{\Omega_0}{2} - \frac{1}{2}\gamma_{\mp}\Omega_{AB}^{\mp}]^2} + \frac{(1-\cos\Theta_{\mp})(1-\chi_{AB}^{\mp})}{[\frac{1}{2}\gamma_{\mp}(1-\chi_{AB}^{\mp})]^2 + [\tilde{\omega} \pm \frac{\Omega_0}{2} + \frac{1}{2}\gamma_{\mp}\Omega_{AB}^{\mp}]^2}\right], \quad (3)$$

where the symbol $\sum_{\mp}$ indicates that both sets of lines appearing with the minus sign and with the plus sign need to be incorporated in the sum. Here $\tilde{\omega} = \omega - \omega_{21}$, $\Theta_{\mp} = 2\pi\frac{r_{AB}}{\lambda_{21}}(1 \mp \frac{\Omega_0}{2\omega_{21}})\cos\theta$ with θ being the angle between the direction of observation and the line connecting the atoms, and $r_{AB} = |\vec{r}_{AB}|$. λ_{21} (ω_{21}) denotes the wavelength (frequency) of the bare atomic transition $|2\rangle \to |1\rangle$ and $\Phi(\vec{r}) = \frac{3}{8\pi}\sin^2\phi$, with ϕ being the angle between the observation direction and the atomic transition dipole moment $\vec{d}_{21}$. $\gamma_{\mp} = \frac{1}{2}\gamma_{21}(1 \mp \frac{\Omega_0}{2\omega_{21}})^3$ represent, respectively, the single-atom spontaneous decay rates from the atomic dressed states $|-\rangle$ and $|+\rangle$, and $\gamma_{21} = 4d_{21}^2\omega_{21}^3/(3\hbar c^3)$ is the free-space spontaneous rate corresponding to the decay of bare state $|2\rangle$ in the absence of the external coherent field. The explicit form of the collective parameters $\chi_{AB}^{\pm} \equiv \chi_{AB}(k_\pm)$ and $\Omega_{AB}^{\pm} \equiv \Omega_{AB}(k_\pm)$ are given by the expressions

$$\chi_{ij}(k_\pm) = \frac{3}{2}\left\{[1-\cos^2\xi]\frac{\sin(k_\pm r_{ij})}{k_\pm r_{ij}} + [1-3\cos^2\xi]\left[\frac{\cos(k_\pm r_{ij})}{(k_\pm r_{ij})^2} - \frac{\sin(k_\pm r_{ij})}{(k_\pm r_{ij})^3}\right]\right\}, \quad (4)$$

$$\Omega_{ij}(k_\pm) = \frac{3}{2}\left\{[\cos^2\xi - 1]\frac{\cos(k_\pm r_{ij})}{k_\pm r_{ij}} + [1-3\cos^2\xi]\left[\frac{\sin(k_\pm r_{ij})}{(k_\pm r_{ij})^2} + \frac{\cos(k_\pm r_{ij})}{(k_\pm r_{ij})^3}\right]\right\}, \quad (5)$$

with $k_\pm = \frac{2\pi}{\lambda_{21}}[1 \pm \Omega_0/(2\omega_{21})]$, and $i \neq j \in \{A, B\}$.

It should be mentioned here that the collective parameters $\chi_{AB}^{\pm}$ and $\Omega_{AB}^{\pm}$ depend critically on the interatomic separation r_{AB}, the ratio $\Omega_0/(2\omega_{21})$, as well as on the angle ξ between the dipole moments $\vec{d}_{21}$ and $\vec{r}_{AB}$. Both parameters tend to zero in the case $k_\pm r_{AB} \to \infty$, which corresponds to the absence of coupling among the emitters. When $k_\pm r_{AB} \to 0$ the parameter $\chi_{AB}^{\pm}$ tends to unity (maximal correlations), while $\Omega_{AB}^{\pm}$ tends to the usual static dipole-dipole interaction potential. The dependence of collective parameters $\chi_{AB}^{\pm}$ and $\Omega_{AB}^{\pm}$ on the ratio $\Omega_0/(2\omega_{21})$ can be understood in the following way. Since we employ strong external fields, the Rabi frequency Ω_0 can be of the order of the atomic transition frequency ω_{21}, i.e., $\Omega_0 \approx \omega_{21} \ll \omega_{32}$. In the dressed states representation [Fig. 1(b)] this means that the $|-\rangle$ state approaches the unperturbed atomic state $|\tilde{1}\rangle$, while the other dressed state $|+\rangle$ moves away from $|\tilde{1}\rangle$. Thus, in keeping the atoms at fixed interatomic separations and changing the transition frequencies, and thus the dipoles involving the dressed states $|\pm\rangle$ and $|\tilde{1}\rangle$, we are able to modify indirectly and substantially the interaction among the radiators.

The spontaneous emission spectrum from all excited states to the ground level $|\tilde{1}_A, \tilde{1}_B\rangle$ is depicted in Fig. 2. It consists of four lines associated with decays from the four collective dressed atomic states $|\Psi_-^{\pm}\rangle = \{|-_A, \tilde{1}_B\rangle \pm |-_B, \tilde{1}_A\rangle\}/\sqrt{2}$ and $|\Psi_+^{\pm}\rangle = \{|+_A, \tilde{1}_B\rangle \pm |+_B, \tilde{1}_A\rangle\}/\sqrt{2}$. As visible from Fig. 1(c) and in agreement with the positions of the four lines in Fig. 2, the two upper sets of dressed states are separated by $\hbar\Omega_0$ via the laser field, and the upper and lower doublets are split each by the dipole-dipole interaction energy $\hbar\gamma_+\Omega_{AB}^+$ and $\hbar\gamma_-\Omega_{AB}^-$, respectively. Equation (3) and Fig. 3(a) demonstrate further that the four decay rates $\Gamma_-^{\pm} = \gamma_-(1 \pm \chi_{AB}^-)$ and $\Gamma_+^{\pm} = \gamma_+(1 \pm \chi_{AB}^+)$ correspond to the decay from the collective atomic states $|\Psi_-^{\pm}\rangle$ and $|\Psi_+^{\pm}\rangle$ and to the widths of the lines in Fig. 2. We note that the long-time evolution of the system is controllable by the applied external field. In particular, for small values of the ratio $\Omega_0/(2\omega_{21}) \ll 1$, the atomic populations in the symmetrical ($|\Psi_+^+\rangle$, $|\Psi_-^+\rangle$) and antisymmetrical ($|\Psi_+^-\rangle$, $|\Psi_-^-\rangle$) atomic states are found to decay with similar probabilities. With increasing ratio $\Omega_0/(2\omega_{21})$, however, the linewidths and decay rates change considerably. The narrowest line in the spectrum corresponds to the decay from the collective antisymmetrical state $|\Psi_-^-\rangle$. As the Rabi frequency $\Omega_0/2$ approaches the atomic transition frequency ω_{21}, $\Gamma_-^- \to 0$ and the Lorentz profile of this line tends towards a delta function. Thus, while keeping the atoms at fixed interatomic separations and increasing the ratio $\Omega_0/(2\omega_{21})$, the collective atomic state $|\Psi_-^-\rangle$ decouples from the interaction with the environmental EMF. This dramatic effect arises especially from the enormous decrease of the modal density with the eigenenergy of $|\Psi_-^-\rangle$ approaching that of the ground state $|\Psi_g\rangle = |\tilde{1}_A, \tilde{1}_B\rangle$ (see a similar effect in a one-atom treatment in [1]). The feasibility of laser control of collective interactions is also visible from Fig. 3(b), where the dipole-dipole interaction energy shows a clear dependence on the laser Rabi frequency $\Omega_0/(2\omega_{21})$.

It should be noted at this point that in general all four collective dressed atomic states $|\Psi_-^{\pm}\rangle$ and $|\Psi_+^{\pm}\rangle$ are excited by the laser field; i.e., in principle only a fraction may be decoupled from the environment. Consequently, we propose in what follows a method to completely transfer all populations into the possibly nondecaying collective state $|\Psi_-^-\rangle$. For this purpose, we introduce incoherent pumping r_p (see Fig. 1) from $|\tilde{1}\rangle$ to $|-\rangle$, which may be realized via a further auxiliary state in energy between $|2\rangle$ and $|-\rangle$. Furthermore, we consider laser Rabi frequencies approaching the atomic transition frequency ω_{21}, forcing us not to neglect nonsecular terms in the dressed equations of motion in the interaction picture anymore.

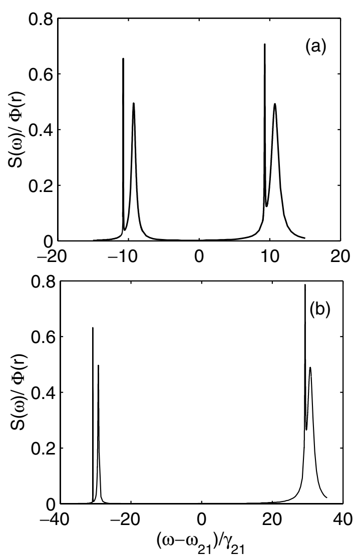

FIG. 2. The collective spontaneous emission spectrum as a function of the frequency of the emitted light ω for $r_{AB}/\lambda_{21} = 0.12$, $\xi = \pi/2$, $\theta = \pi/4$, and $r_p = 0$. Further, for (a) $\Omega_0/(2\omega_{21}) = 0.1$, $\Omega_0/(2\gamma_{21}) = 10$, and for (b) $\Omega_0/(2\omega_{21}) = 0.3$, $\Omega_0/(2\gamma_{21}) = 30$.

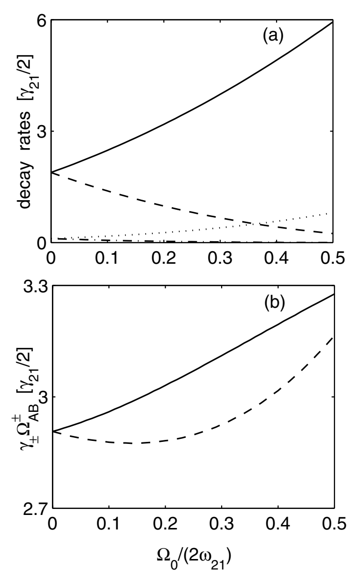

FIG. 3. (a) The dependence of collective spontaneous emission rates as a function of the ratio $\Omega_0/(2\omega_{21})$. The solid (dashed) line corresponds to the decay Γ_+^+ (Γ_-^+) from the symmetrical states $|\Psi_+^+\rangle$ ($|\Psi_-^+\rangle$), while the dotted (dash-dotted) line corresponds to the decay Γ_+^- (Γ_-^-) from the antisymmetrical states $|\Psi_+^-\rangle$ ($|\Psi_-^-\rangle$). (b) The dependence of the dipole-dipole interaction energies $\gamma_-\Omega_{AB}^-$ (solid line) and $\gamma_+\Omega_{AB}^+$ (dashed line) as a function of $\Omega_0/(2\omega_{21})$. In both cases $rAB/\lambda 21 = 0.12$,ξ=π/2\hbox{\curr,} $r_p = 0$.

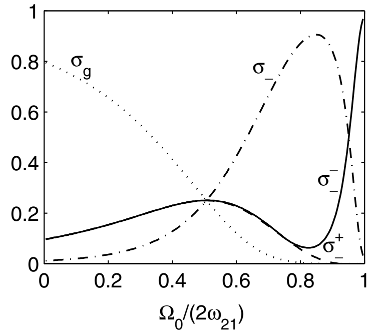

FIG. 4. The population of the collective two-atom states as a function of $\Omega_0/(2\omega_{21})$ for $r_{AB}/\lambda_{21} = 0.7$, $\xi = \pi/2$, $r_p = 0.12$, and $\omega_{21} = 400$ scaled in units of $\gamma_{21} = 1$. The solid (dashed) line corresponds to the population σ_-^- (σ_-^+) of the state $|\Psi_-^-\rangle$ ($|\Psi_-^+\rangle$), while the dotted (dash-dotted) line depicts the population σ_g (σ_-) of the state $|\Psi_g\rangle$ ($|\Psi_-\rangle$).

The additional terms introduce the transitions $|+\rangle \to |\tilde{1}\rangle \to |-\rangle$ and vice versa. The steady-state values of all small parameters of interest are then evaluated to first order in $\gamma_\pm$, r_p, $\gamma_\pm \chi_{AB}^\pm$, and $\gamma_\pm \Omega_{AB}^\pm$ with respect to Ω_0 and then inserted into the remaining equations describing the dynamics of the dressed populations. Figure 4 displays the steady-state results in this approximation. Because of the incoherent pumping into the $|-\rangle$ state, the collective atomic states $|\Psi_+^+\rangle$ and $|\Psi_+^-\rangle$ become unpopulated. For small values of $\Omega_0/(2\omega_{21})$, almost all population remains in the ground state $|\Psi_g\rangle \equiv |\tilde{1}_A, \tilde{1}_B\rangle$. In raising the ratio $\Omega_0/(2\omega_{21})$, the ground state depopulates because in this case the remaining collective spontaneous decay rates of interest from $|\Psi_-^\pm\rangle$ and $|\Psi_-\rangle \equiv |-_A, -_B\rangle$ eventually become smaller than the incoherent pumping rate. Once $\Omega_0/(2\omega_{21})$ reaches values between 0.8 and 0.9, the decay rates from the collective atomic states $|\Psi_-\rangle$ and $|\Psi_-^+\rangle$ become approximately equal, i.e., $\Gamma_- \approx \Gamma_-^+$. However, the decay rate from the antisymmetrical collective atomic state $|\Psi_-^-\rangle$ (Γ_-^-) is significantly smaller than Γ_- and Γ_-^+, so that eventually $|\Psi_-^-\rangle$ becomes highly populated and the remaining ones empty. This result has proven valid for separations among the emitters of order $0.5 < r_{AB}/\lambda_{21} < 1.5$, and the complete population transfer into state $|\Psi_-^-\rangle$ may be reversed to state $|\Psi_-\rangle$ and $|\Psi_-^+\rangle$ for decreasing driving laser field.

In conclusion, a method for manipulating collective atom-environment interaction by a possibly strong external laser field has been demonstrated. Via the control of the involved vacuum modes due to changing dressed transition frequencies, it was especially shown how to trap population in or how to release it from a collective two-particle system.

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