Subradiance in multiply excited states of dipole-coupled V-type atoms

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Abstract – We generalize the theoretical modeling of collective atomic super- and subradiance to the multilevel case including spontaneous emission from several excited states towards a common ground state. We show that in a closely packed ensemble of $N$ atoms with $N-1$ distinct excited states each, one can find a new class of non-radiating dark states, which allows for long-term storage of $N-1$ photonic excitations. Via dipole-dipole coupling only a single atom in the ground state is sufficient in order to suppress the decay of all $N-1$ other atoms. By means of some generic geometric configurations, like a triangle of V-type atoms or a chain of atoms with a $J=1 \rightarrow J=0$ transition, we study such subradiance including dipole-dipole interactions and show that even at finite distances long lifetimes can be observed. While generally hard to prepare deterministically, we identify various possibilities for a probabilistic preparation via a phase-controlled laser pump and decay.

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Introduction. – Quantum fluctuations in the electromagnetic vacuum field inevitably lead to energy dissipation from excited atomic states via the spontaneous emergence of photons [1] known as spontaneous emission. In a quantum electrodynamics treatment the probability for this process and its corresponding decay rate $\Gamma = \omega_0^3/3(\pi\epsilon_0\bar{c}/3)^3$ was first derived by Weiskopf and Wigner [2]. It is proportional to the third power of the transition energy between the excited and lower-lying state as well as to the square of the transition dipole moment between those two states.

As there is only one electromagnetic vacuum, atoms in close proximity will experience correlated fluctuations inducing cooperative effects in their dissipative behavior. By means of constructive as well as destructive interference of the emerging photons the collective spontaneous emission rates are drastically modified as a function of distance [3–6]. A strongly increased spontaneous emission is dubbed “superradiance” while a decreased rate is referred to as “subradiance” [7].

Due to the quantum nature of atomic excitations, they can be delocalized and distributed over an entire atomic ensemble, exhibiting highly multi-partite entanglement [8–10]. Well-known examples are the single-excitation Bell states of two atoms [11,12], the W-state [13,14] and many others, e.g., [15].

Depending on the geometry of the atomic ensemble as well as on the local phase difference of the excitation amplitudes between the atoms [16], such delocalized excitation states can feature either super- or subradiance. For instance, for two closely spaced atoms ($d \ll \lambda_0 = 2\pi c/\omega_0$), the symmetric Bell state $|+\rangle = (|eg\rangle + |ge\rangle)/\sqrt{2}$ is superradiant, while its asymmetric analogue $|-\rangle = (|eg\rangle - |ge\rangle)/\sqrt{2}$ is strongly subradiant and decouples from the radiation field completely at distances close to zero [17]. This leads to the term “dark state”. Because of the fact that their lifetime is often orders of magnitude longer than typical experimental cycles, those dark states are a valuable resource in quantum information storage and processing [18–20].

While subradiant states of dense atomic ensembles are easy to identify theoretically [15,21–23], they have been quite elusive and hard to find in concrete experiments [24,25], with directional emission patterns as one of the signatures of destructive interference leading to subradiance [26]. Besides the influence of motion and various dephasing mechanisms, it was recently pointed out, that the complex level structure of typical atoms beyond...
A two-level approximation will often prevent the appearance of perfectly dark states [27]. In particular, for excited atomic states, which can decay to different lower states via more than one decay channel, the observation of subradiance is much more challenging. It can be easily shown that for a system of two Λ-type atoms no dark state can be found, as both decay channels need to be blocked via interference, which cannot be achieved simultaneously. However, in earlier work [27] we could show that an ensemble of N - 1 excited states |e⟩ with independent transitions to a common ground state |g⟩, represented by σj. The collective decay rates are given by ΓN j j ′, whereas the collective energy shifts are written as ΩN k j j ′. The individual spontaneous emission rate for transition j in all atoms is γj.

Model. – Even though the model we are introducing here allows for an arbitrary number of atoms each with an arbitrary number of excited states with one common ground state, in our investigations below we will focus on level structures involving two and three upper levels as our aim is to show the possibility of subradiance in emitters with more than one upper level. Experimentally, such a level scheme can be addressed with π- and σ± polarized laser light on the J = 0 → J = 1 transition. More specifically, the ground state might be 1S0 and the excited states 3P m with m ∈ {±1, 0} in 88Sr.

Let us now consider a collection of N identical multi-level atoms at fixed positions {r j}N j=1. Each atom features N - 1 excited states {|e j⟩}N j=1 at energies ωj with dipole coupling to a common ground state |g⟩ via a transition dipole moment of μ j.

The combined Hamiltonian of the atoms and the electric field is given by

$$H = H_A + H_F + H_{\text{int}},$$

with the atomic part

$$H_A = \sum_{i=1}^{N} \sum_{j=1}^{N-1} \omega_j \sigma_j^+ \sigma_j^-$$

and the field

$$H_F = \sum_{\vec{k},\lambda} \omega_{\vec{k},\lambda} a_{\vec{k},\lambda}^\dagger a_{\vec{k},\lambda}.$$

The interaction between the atoms and the field in dipole approximation is then

$$H_{\text{int}} = -\sum_{i=1}^{N} \sum_{j=1}^{N-1} (\mu_i^\dagger \sigma_j^+ \cdot \vec{E}(\vec{r}_i) + \text{h.c.}),$$

where \(\vec{E}(\vec{r}_i)\) is the quantized electric field. When particularizing to N = 3 below, we will consider a situation where the transition dipole matrix elements inside each atom are mutually orthogonal and real, that is

$$\mu_j^\dagger \cdot \mu_j = 0.$$

After tracing out the electromagnetic field modes in a standard quantum optics fashion assuming the field in its vacuum state [6,29–31] the system dynamics can be described by the master equation

$$\dot{\rho} = i[\rho, H] + \mathcal{L}[\rho],$$

with the effective Hamiltonian including dipole-dipole interaction

$$H = \sum_{i=1}^{N} \sum_{j=1}^{N-1} \omega_j \sigma_j^+ \sigma_j^- + \sum_{i \neq k} \sum_{j,j'} \Omega_{ij}^{kk} \sigma_j^+ \sigma_{j'}^-, \sigma_j^- \rho$$

and the Liouvillian in Lindblad form

$$\mathcal{L}[\rho] = \sum_{i,k} \sum_{j,j'} \Gamma_{ij}^{kk}(\rho \sigma_j^+ \rho^\dagger + \sigma_j^+ \rho \sigma_j^- - \sigma_j^+ \sigma_j^- \rho - \rho \sigma_j^+ \sigma_j^-),$$

where \(\sigma_j^+\) denotes the rising (lowering) operator of the j-th transition in the i-th atom.
The coherent part of the dipole-dipole interaction induces energy shifts (see fig. 1) due to the couplings

$$\Omega_{jj'}^{ik} = \frac{3\sqrt{\lambda_j \lambda_{j'}}}{2} \left[ (\hat{\mu}_i^\dagger \cdot \hat{\mu}_{j'}^\dagger) P_R(k_0 r_{ik}) - (\hat{\mu}_j^\dagger \cdot \hat{\mu}_{j'}^\dagger) P_I(k_0 r_{ik}) \right],$$

while the incoherent collective dissipation is characterized by

$$\Gamma_{jj'}^{ik} = \frac{3\sqrt{\lambda_j \lambda_{j'}}}{2} \left[ (\hat{\mu}_i^\dagger \cdot \hat{\mu}_{j'}^\dagger) Q_R(k_0 r_{ik}) - (\hat{\mu}_j^\dagger \cdot \hat{\mu}_{j'}^\dagger) Q_I(k_0 r_{ik}) \right].$$

Furthermore, for brevity we have introduced the functions

$$P_R(\xi) = \frac{\cos \xi - \xi \sin \xi}{\xi^2 - 3 \cos \xi},$$

$$P_I(\xi) = \frac{\sin \xi - \xi \cos \xi}{\xi^2 - 3 \cos \xi},$$

$$Q_R(\xi) = \frac{\cos \xi}{\xi^2} - 3 \sin \xi - 3 \cos \xi,$$

$$Q_I(\xi) = \frac{\sin \xi}{\xi^2} + 3 \cos \xi - 3 \sin \xi,$$

where $r_{ik} = |\vec{r}_i - \vec{r}_k|$ represents the interatomic distance between atom $i$ and atom $k$, and $k_0 = \omega_0/c$ with $\omega_0 = (\omega_j + \omega_{j'})/2$ and $\Gamma_{jj'}^{ik} = \gamma_j = 2\mu_j^2 \omega_j^3/(3\epsilon_0 c^3)$ is the spontaneous emission rate of a single atom on the $j$-th transition. The couplings for the energy shifts as well as the collective decays are plotted in fig. 2 as a function of the interatomic distance, whereas varying the dipole moment orientations leads to oscillations of various amplitudes (see fig. 3). The terms $\Omega_{12}^{12}$ and $\Omega_{12}^{12}$ are dipole-dipole cross-coupling coefficients, which couple dipoles even though they are orthogonal [6].

**Equilateral triangle: analytical treatment.** – We begin by analyzing an analytically accessible configuration where the three emitters are positioned in an equilateral triangle. Later on, we will numerically investigate a linear chain of emitters, which is a more feasible setup from an experimental point of view.

For three 3-level atoms placed at the corners of an equilateral triangle with dipole orientations chosen such that the configuration features a C3 symmetry (see fig. 3), the states $|\Psi_{sr}^3\rangle$ and $|\Psi_{sr}^3\rangle$ are both eigenstates of the Hamiltonian from eq. (6) whose energies can be calculated explicitly. For three V-type atoms $|\Psi_{d}^3\rangle$ is given by

$$|\Psi_{d}^3\rangle = \frac{1}{\sqrt{6}} \left( |e_1 e_2 g\rangle + |g e_1 e_2\rangle + |e_2 g e_1\rangle - |e_1 g e_2\rangle - |e_2 e_1 g\rangle - |g e_2 e_1\rangle \right),$$

whereas in the superradiant analogue $|\Psi_{sr}^3\rangle$, which is comprised of the exact same bare states, all signs are positive.

Clearly, the dynamics of any eigenstate of the Hamiltonian is restricted to the decay towards other eigenstates $|\psi_{eig}\rangle$ induced by the Liouvillian, i.e., $\dot{\rho}_{eig} = \mathcal{L}[\rho_{eig}]$ with $\mathcal{L}[\rho_{eig}] = |\psi_{eig}\rangle \langle \psi_{eig}|$. The corresponding rates can be found by calculating the overlap with all other states. The decay and feeding rates for a certain selection of states are shown in fig. 4. Explicitly, the decay rate for the eigenstate $|\psi_{eig}\rangle$ is given by $\langle \psi_{eig}| \mathcal{L}[\rho_{eig}] |\psi_{eig}\rangle$.

We find that the lowest-lying energy state in the double-excitation manifold corresponds to the antisymmetric dark state $|\Psi_{d}^3\rangle$, while the highest-energy state is the superradiant state. With a more and more pronounced subradiance in $|\Psi_{d}^3\rangle$ at decreasing interatomic distances, also its feeding rate from higher-lying states decreases, which culminates in a decoupling from all other states and the electromagnetic field. In particular, for the equilateral triangle configuration, the lower an eigenstate lies energetically, the smaller its decay rate is, as can be seen for selected states in fig. 4. A full account of all coupling and feeding rates is available in the Supplementary Material Supplementarymaterial.pdf (SM). Also note that all feeding and decay rates to and from a particular state sum up to zero.

**Numerical diagonalization for up to four atoms.** – We now analyze the scaling of the decay rates as a function of the interatomic distance for increasing atom numbers and different geometries.

In fig. 5(a) the simple case of two two-level atoms is shown, where the sub- and superradiant decay rates oscillate around the independent decay rate $\Gamma$ with an amplitude decreasing with the interatomic distance, such that...
Fig. 4: Decay cascade. After diagonalizing the Hamiltonian for the equilateral triangle configuration of three 3-level atoms with symmetric dipole orientations we show the decay cascade for selected eigenstates (the full cascade can be found in the SM). We define $\gamma_1 \equiv \frac{\Gamma}{2}\Gamma_{ij}(kr)\, (k\neq r)$ and $\gamma_2 \equiv -\frac{\Gamma}{2}\Gamma_{ij}(kr) + \frac{\Gamma}{2}\Gamma_{ij}(r\neq k)$ and $\Gamma$ represents the spontaneous emission rate of a single V-type atom with degenerate excited states. Additionally, the collective energy shifts in the respective excitation manifolds are shown with $\Omega_k \equiv \Omega_k^{ij\,r}$ and $\omega_0$ being the energy between ground and excited states.

Fig. 5: Decay rates in regular arrays. (a) Sub- and superradiant decay rates for two dipoles as a function of the interatomic distance, where $\gamma = \frac{\Gamma}{2}\Gamma_{ij}(kr)$ and the black dashed line indicates the lowest decay rate at any given distance. (b) Lowest decay rates in the $(N - 1)$-excitation manifold for $N$ $N$-level emitters as a function of the interatomic distance, with the white circles corresponding to two 2-level atoms, the red circles to three 3-level V-type atoms in a linear chain, the orange triangles to an equilateral triangle, the blue circles to four 4-level V-type atoms in a chain and the white squares to four atoms in a square.

Fig. 6: Lowest decay rates. The lowest decay rates (a) for three 3-level V-type atoms in a triangle configuration for eigenstates in the two-excitation manifold for different distances between atoms 1, 2 and atoms 1, 3, respectively, and (b) in a chain of atoms for different distances between atoms 1, 2 and atoms 2, 3 where all transitions are orthogonal to the direction of the chain are shown.

the super- and subradiant state switch their roles at each node. The black dashed curve corresponds to the lowest decay rate at any given distance and is generalized to more involved configurations in fig. 5(b).

In fig. 5(b) it can be seen, that the lowest collective decay rate for the $(N - 1)$-excitation manifold for $N$ atoms goes to zero only if the interatomic distances approach zero, if all dipole transition moments are orthogonal to the plane of the atomic ensemble. For the equilateral triangle with symmetric dipole orientations and for $N \geq 4$ atoms with more than two transitions this is not possible anymore and the minimal decay rate is (see fig. 6).

**Dark state preparation.** — In most geometric configurations apart from the equilateral triangle the antisymmetric state $|\psi_{d}^{3}\rangle$ is not an exact eigenstate of the Hamiltonian from eq. (6). Yet, its subradiant property will prevail as shown for a linear chain in fig. 7. The state $|\psi_{\text{app}}\rangle = (|e_{1} e_{2}\rangle + |e_{2} e_{3}\rangle)/\sqrt{2}$ denotes a product state with atoms 1 and 2 entangled and exhibits subradiance as well. Generally, subradiance becomes particularly apparent at small atomic distances, where the derivative of the incoherent coupling with respect to $r$ is almost zero.

At finite distances $|\psi_{d}^{3}\rangle$ can couple to other states and will therefore decay as shown in fig. 7. Naturally, this means that it can be populated via decay from a higher-lying state, which in this case are all triply excited states. A typical case where the dark state becomes populated by photon emission for a three qutrit chain prepared in a totally inverted state, $|e_{1} e_{2} e_{3}\rangle$, is demonstrated in fig. 8. Note that there are, in fact, eight different possibilities for triply excited states, i.e., $|e_{i} e_{j} e_{k}\rangle$ with $i,j,k \in \{1,2\}$, which lead to similar results. In fig. 8 it can be seen, that the dark state can acquire a significant population, even via purely dissipative preparation, by choosing an appropriate geometric configuration. On the other hand, the feeding rate for the dark state becomes smaller with decreasing distances as it starts to decouple from the electromagnetic field.

As we have seen above, after an initial build-up of population in the dark state, the remainder of the population...
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most likely ends up in the ground state. Hence, one can think of reusing the atoms in the ground state in order to further increase the occupation of the dark state. For this purpose, the preparation of the dark state $|\Psi_d^3\rangle$ or its superradiant analogue $|\Psi_{sr}^3\rangle$ can be facilitated by a continuous pump laser. It turns out that using different excitation phases for each atom can strongly improve the efficiency of this process, although this might be challenging to implement in practice.

We include a continuous pump in our model by adding the term $H_{\text{pump}} = \sum_{i=1}^{3} \sum_{j=1}^{2} \eta_i (\sigma_j^+ + \sigma_j^-)$ to the Hamiltonian with $\eta_1 = \eta$, $\eta_2 = \eta e^{i\varphi_1}$ and $\eta_3 = \eta e^{i\varphi_2}$, assuming that all atoms are driven with the same strength $\eta$.

In our example the atoms are initialized in the ground state, $|g,g,g\rangle$, and we look at the population of the dark state after a given laser illumination time.

In fig. 9 the preparation probabilities for $|\Psi_d^3\rangle$ in a linear chain and for its superradiant analogue $|\Psi_{sr}^3\rangle$ in an equilateral triangle are shown as a function of the laser phase using a constant pump amplitude of $\eta = 8.5\Gamma$ and laser phases $\varphi_1 = \pi/3$, $\varphi_2 = \varphi_3 = \pi/3$, assuming that all atoms are driven with the same strength $\eta$.

Now, we include different phases for different transitions by writing our pump Hamiltonian as $H_{\text{pump}} = \sum_{i=1}^{3} \sum_{j=1}^{2} \eta_i (\sigma_j^+ + \sigma_j^-)$. Figure 10(a) shows the...
preparation probability for $|\Psi^3_3\rangle$ for a range of different phases, where for instance for $\varphi_1 = \varphi_2 = 7/10\pi$ a maximum of $\approx 24\%$ is reached after $\Gamma t = 0.3$. In fig. 10(b) we compare the time evolution of a pulsed laser with a continuous drive. Both cases lead to the same maximal value after $\Gamma t = 0.3$, but, after turning off the laser the dissipative dynamics lead to larger preparation probabilities shortly after that. Only for times longer than $\Gamma t = 1.5$ the laser driven system dominates the preparation probability. Specifically, for the case of pulsed lasing in fig. 10(b) the first peak corresponds to a preparation probability of 24% and the second peak to 15%, both within an evolution time of $\Gamma t = 1$.

Conclusions. – We have generalized the concept of subradiance to multilevel emitters with several excited atomic levels decaying via independent decay channels towards a common ground state. In these systems the most subradiant states are completely anti-symmetric and maximally entangled. In contrast to ensembles of two-level emitters this multilevel type of dark states can hold several excitation quanta without decay. Hence detection could be facilitated by non-classical photon correlations at long time delays. An experiment for pumping V-type atoms and subsequent detection of super- or subradiance via resonance fluorescence was done in [32] and their setup could be used to implement our proposed driving scheme. Also a very recent idea for preparing dark states of fermionic emitters in optical lattices, each featuring multiple energy levels, was proposed in [33] and constitutes a realistic platform for our suggested system.

Entangled subradiant states have promising applications in quantum information processing and optical lattice clocks [34,35], amongst other key quantum technologies, where longer coherence times and a better understanding of energy level shifts induced via dipole-dipole interactions are crucial for improved accuracies.

States that do not decay as they decouple from the radiation field in turn are hard to access in order to prepare them directly. Yet, a probabilistic preparation can be achieved via spontaneous emission from higher-lying states or in a much more efficient way by the application of laser pulses with spatial phase control.

Future work in this line of studies will include coupling to a cavity field and analyzing the emission and absorption behaviour of multiple V-type emitters via an input/output formalism as in [36]. Another direction is to investigate waveguiding effects within the above model in the second excitation manifold as was done for single excitations in [37] for two-level emitters and for emitters with multiple levels in the ground and excited state as in [38].

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