Subradiance via Entanglement in Atoms with Several Independent Decay Channels

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Spontaneous emission of atoms in free space is modified by the presence of other atoms in close vicinity inducing collective super- and subradiance. For two nearby atoms with a single decay channel the entangled antisymmetric superposition state of the two single excited states will not decay spontaneously. No such excited two-atom dark state exists, if the excited state has two independent optical decay channels of different frequencies or polarizations. However, we show that for an excited atomic state with $N-1$ independent spontaneous decay channels one can find a highly entangled N-particle dark state, which completely decouples from the vacuum radiation field. It does not decay spontaneously, nor will it absorb resonant laser light. Mathematically, we see that this state is the only such state orthogonal to the subspace spanned by the atomic ground states. Moreover, by means of generic numerical examples we demonstrate that the subradiant behavior largely survives at finite atomic distances including dipole-dipole interactions.

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Spontaneous decay of an excited atomic state towards lower lying states via optical photon emission is a striking consequence of the quantum nature of the radiation field [1]. Introduced even before Einstein, the spontaneous emission rate $\Gamma = \omega^3\mu^2/3\epsilon_0\pi\hbar c^3$, now called Einstein's A coefficient, is proportional to the squared transition dipole moment μ^2 between the upper and lower atomic state and the third power of the transition frequency ω [2].

For several particles in close vicinity the emission process is not independent but enhanced or reduced collectively, depending on the emitters geometry [3,4]. These superradiant and subradiant collective states, where a single excitation is distributed over many particles, are entangled states [5,6]. Although a recent classical coupled dipole model leads to subradiancelike phenomena as well [7], the most superradiant and the perfect dark states for two two-level quantum emitters with states $(|g\rangle, |e\rangle)$ are the maximally entangled symmetric and antisymmetric superposition states,

$$|\psi_\pm\rangle = (|eg\rangle \pm |ge\rangle)/\sqrt{2}. \tag{1}$$

While superradiance on a chosen transition persists when the atom possesses more than one decay channel, no completely dark state for two atoms with several decay paths from an excited state $|e\rangle$ to a couple of lower lying states $|g_i\rangle$, as depicted in Fig. 1, is known. Since there are alternative possibilities for decay in most atomic systems, observing subradiance experimentally proves much more difficult than seeing superradiance, as all decay channels need to be blocked [8–11].

In this Letter, we introduce a generalized class of dark or subradiant states for atoms with several independent transitions. As a key result of this work we find that for systems of N particles highly entangled multipartite states, where $N-1$ independent decay channels are suppressed, exist. For these states the total dipole moments on all $N-1$ transitions vanish simultaneously and, at least in principle, any optical excitation in this state can be stored indefinitely. Note that subradiance of multilevel atoms has been studied before but decay was largely limited to a single degenerate channel for all atoms and transitions [10].

After having introduced our model and the generalized unique multipartite entangled dark states, we will discuss the relation between subradiance and their special entanglement properties as well as possible quantum information theoretical procedures to prepare them. Their mathematical properties are detailed in the Supplemental Material [12]. In the final part of the Letter we study subradiance for some generic examples of three-level Λ-atoms, including dipole-dipole coupling, where population of the dark state can be accumulated via decay from multiply excited states.

Interestingly, a related phenomenon appears in V-type atoms with two excited and one ground state, where a single ground state atom can prevent decay of several excitations as exhibited in the Supplemental Material [12].

Model.—Let us assume a collection of N identical N-level emitters with a set of $N-1$ low energy eigenstates

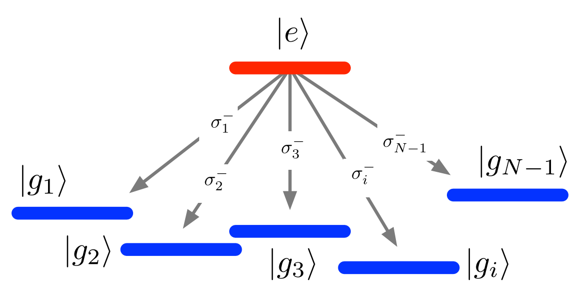

FIG. 1. Level scheme of an atom with several independent decay channels of different polarization or frequency.

$|g_i\rangle$, where $i \in \{1, ..., N\}$, which are dipole coupled to a higher energy state $|e\rangle$ separated by the excitation energies $\hbar\omega_i$ (see Fig. 1). The atomic center of mass motion is treated classically with fixed positions $\mathbf{r}_i$ within a cubic wavelength. For each atom i and transition j we define individual Pauli ladder operators $\sigma_j^{i\pm}$ describing transitions between the ith atom's excited state $|e\rangle_i$ and its j lower energy states $|g_j\rangle_i$, respectively.

The coupling of each atomic transition j of atom i, to the electromagnetic vacuum leads to an individual free space decay rate Γ_j^i. As all atoms are coupled to the same vacuum modes, these decay rates are modified by pairwise interactions with neighboring atomic transitions k, l, which upon elimination of the field modes can be described by mutual decay rates Γ_{jl}^{ik}, with $\Gamma_j^{ii} = \Gamma_j$ [3,18]. Note, that in addition to the modified decay properties the collective vacuum coupling induces resonant energy exchange terms Ω_{jl}^{ik} as presented in Refs. [3,4,18,19].

As our central interest is the modification of the collective emission rates, for simplicity, we will assume a highly symmetric arrangement of the particles, so that all particles acquire equal energy shifts, i.e., $\Omega_{jj}^{ik} = \Omega_j$, which can be incorporated into effective transition frequencies [19]. In terms of the operators defined above with the excited state energy set to zero the dipole coupled atomic Hamiltonian is given by

$$H = \sum_{i,j} -\bar{\omega}_j^i \sigma_j^{i-} \sigma_j^{i+} + \sum_{i \neq k} \sum_j \Omega_j^{ik} \sigma_j^{i+} \sigma_j^{k-}. \quad (2)$$

The full dynamics of the coupled open system including decay is governed by a master equation for the density matrix ρ of the whole system of N multilevel emitters,

$$\frac{\partial \rho}{\partial t} = i[\rho, H] + \mathcal{L}[\rho]. \quad (3)$$

Following standard quantum optical assumptions and methods the effective Liouvillian summed over all transitions and atom pairs reads [19,20]

$$\mathcal{L}[\rho] = \frac{1}{2} \sum_{i,k,j} \Gamma_j^{ik} [2\sigma_j^{i-} \rho \sigma_j^{k+} - \sigma_j^{i+} \sigma_j^{k-} \rho - \rho \sigma_j^{i+} \sigma_j^{k-}]. \quad (4)$$

While this can be a complex and complicated expression for a general arrangement [19], in the case of atomic distances much smaller than the transition wavelength, all $\Gamma_j^{ik} = \Gamma_j$ become independent of the atomic indexes (i, k), reducing to a single constant Γ_j. For simplicity, we also assume equal decay rates on all transitions $\Gamma_j = \Gamma$, i.e., equal dipole moments and Clebsch-Gordan coefficients [21]. This will hardly be exactly true for any real atomic configuration (besides $J = 0$ to $J = 1$), but it will not change the essential conclusions below.

Collective atomic dark states.—Obviously, any atomic density matrix ρ_g involving ground state populations $|g_i\rangle$ only is stationary under $\mathcal{L}$ in a trivial way with $\mathcal{L}[\rho_g] = 0$. Therefore, states ρ_e featuring atomic excitations, which will still not decay under $\mathcal{L}$, are much more interesting.

For the case of two two-level atoms such dark states are well known and have been confirmed experimentally decades ago [22]. They are antisymmetric superpositions as introduced in Eq. (5), i.e., $|\psi_d^2\rangle = |\psi_-\rangle$. As a central claim of this work we show that this formula can be generalized to the case of N atoms with $N - 1$ independent optical transitions between the upper state $|0\rangle = |s_0\rangle = |e\rangle$ and $N - 1$ lower states $|i\rangle = |s_i\rangle = |g_i\rangle$ in the form

$$|\psi_d^N\rangle = \frac{1}{\sqrt{N!}} \sum_{\pi \in S_N} \mathrm{sgn}(\pi) \bigotimes_i |s_{\pi(i)}\rangle, \quad (5)$$

where the sum runs over all permutations π of N elements. Using the criterion for pure states to be stationary under $\mathcal{L}$ given in Ref. [23], we show in the Supplemental Material [12] that this N-level state of total spin 0 is the unique stationary state orthogonal to the subspace, where all particles are in $|g_i\rangle$ for some i. A symmetric variant of this state, denoted by $|\psi_{\mathrm{sr}}^N\rangle$, with all positive signs will be its superradiant analogue.

The dark state has a zero total dipole moment $\mu_j = \langle \sum_i \sigma_j^i \rangle = 0$ on any transition as a consequence of its symmetry. This implies strong entanglement as discussed in more detail below. Indeed, those states are a special case of complex entangled states, many of whose mathematical properties have been considered before (see, e.g., Ref. [24]).

For three Λ atoms one explicitly gets

$$|\psi_d^3\rangle = \frac{1}{\sqrt{6}} \{ |e g_1 g_2\rangle + |g_1 g_2 e\rangle + |g_2 e g_1\rangle - |e g_2 g_1\rangle - |g_2 g_1 e\rangle - |g_1 e g_2\rangle \}, \quad (6)$$

which is within the set of maximally entangled tripartite states of qutrits [25].

As mentioned before, this state is stationary in the case of $H = 0$ and coinciding decay rates, $\Gamma_j^{ik} = \Gamma$ (see also Fig. 3). For more realistic situations, in Fig. 2 we consider a subwavelength equilateral triangular configuration with $d = 0.08\lambda$. The subradiant decay resulting from the evolution governed by the master equation, Eq. (3), is shown. As the atoms also experience energy shifts from the resonant dipole-dipole coupling, Ω_j^{ik}, in Eq. (2), the dark states in Eq. (5) will, in general, not be eigenstates of H and dynamic mixing with other states induces a finite lifetime as for two-level dark states [26]. In this graph, in order to demonstrate the subradiance effect more clearly, we have set $\Omega_j^{ij} = 0$.

Note that the subradiant states discussed here are not the dark states appearing in a two-laser excitation of Λ-type systems discussed in Ref. [27]. There, a particular

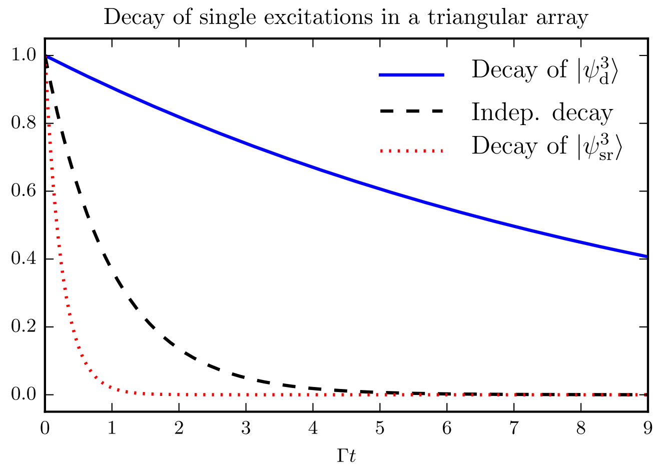

FIG. 2. Upper state population decay of three interacting Λ-type atoms in an equilateral triangle of size $d = 0.08\lambda \ll \lambda$, where $\Gamma_j^{ik} \approx 0.95\Gamma_j$ for all particles i, k with $i \neq k$ starting from the ideal dark state (solid blue line). For comparison, the dashed black line shows independent atom decay, while the dotted red line corresponds to a fully symmetric state with superradiant decay on both transitions. Coherent level shifts are neglected, i.e., $\Omega_j^{ik} = 0$ for all i, k.

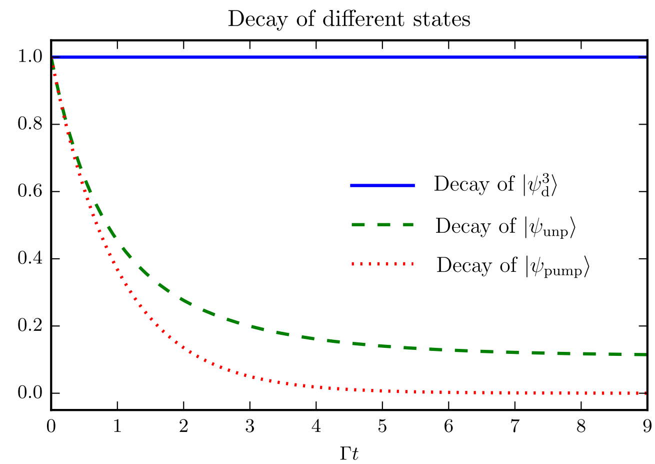

FIG. 3. Upper state population decay for three closely spaced Λ-type atoms for different singly excited initial states, where $\Gamma_j^{ik} = \Gamma$ and we neglect Ω_j^{ik}. The solid blue line corresponds to the dark state $|\psi_d^3\rangle$, the dotted red line gives the case of the two atom dark state for optically pumped atoms $1/\sqrt{2}(|eg_1\rangle - |g_1 e\rangle)|g_1\rangle$ and dashed green refers to an unpolarized product state $1/\sqrt{2}(|eg_1\rangle - |g_1 e\rangle)|g_2\rangle$ involving all three atomic states.

superposition of ground states decouples from the laser excitation for each atom separately and leads to a coherent population trapping in the ground state manifold. The notion of dissipative state preparation [23,28] has also been used in Ref. [29]. There, it has been shown that three M-shaped 5-level atoms interacting via three coupled cavities and lasers can be driven into the state $|\psi_d^3\rangle$, where, however, only atomic ground states are involved.

As an important consequence of the uniqueness of the dark state, no such state can exist for a smaller number of atoms. Therefore, when considering M atoms with $N - 1$ independent optical transitions between the upper state $|e\rangle$ and their $N - 1$ lower states $|g_i\rangle$, where the emitted photons on each transition are distinguishable, the following picture emerges: for $M < N$ only ground states are stationary under $\mathcal{L}$. In case $M > N$, however, extra stationary states involving excitations can be found. They are given by tensor products of states that are stationary for parts of the system and superpositions of these states. To give a simple example for the case $M = 6$ and $N = 3$ the states

$$|\psi\rangle = (\alpha|\psi_d^3\rangle \otimes |\psi_d^3\rangle + \beta|g_i g_j\rangle \otimes |\psi_d^3\rangle \otimes |g_k\rangle)/\sqrt{2} \quad (7)$$

are dark for any $\alpha, \beta \in \mathbb{C}$.

Entanglement properties of dark states.—Important properties of the dark states $|\psi_d^N\rangle$ will shortly be recapitulated here as they provide important insights into the physical origin of subradiance as well as possibilities to prepare them. It has been shown via the construction of generalized Bell inequalities for any N that there exists no local hidden variable model describing their quantum predictions [24]. Hence, $|\psi_d^N\rangle$ has no direct classical analogue. Moreover, it can be used to solve the Byzantine agreement problem, the N strangers problem, the secret sharing problem, and the liar detection problem [24,30].

What makes these states so useful for the above mentioned tasks are their very special entanglement properties [24], which we briefly reiterate here (for more details see Ref. [12]). First, note that the state is contained in the maximally entangled set, which is a generalization of the bipartite maximally entangled state [25]. The bipartite entanglement shared between any of the particles and the rest of the particles is maximal as it is for pairs of two-level dark states. This property implies that the reduced density matrix for any particle, j obtained by the partial trace over all other particles is proportional to identity. Hence, we see that each particle contains no individual information and, in this sense, subradiance is a purely nonlocal, nonclassical phenomenon.

An important property of $|\psi_d^N\rangle$ is the fact that for all invertible operators S, $|\psi_d^N\rangle \propto S^{\otimes N}|\psi_d^N\rangle$. This symmetry has several important consequences. If one particle is measured in any basis and the measurement outcome and chosen basis are announced, the other $N - 1$ particles can be transformed to the state $|\psi_d^{N-1}\rangle$ deterministically by performing local unitary operations only [31]. This implies that one can generate $|\psi_d^N\rangle$ from $|\psi_d^{N-1}\rangle$ with an extra atom, as will be explained below.

Let us note here, that the geometric measure of entanglement [32] can be computed easily and one obtains that $E_g(|\psi_d^N\rangle) = 1 - \max_{|a_1\rangle,\ldots,|a_N\rangle} |\langle a_1, \ldots, a_N|\psi_d^N\rangle|^2 = 1 - (1/N!)$ [12,33]. Furthermore, it can be shown that the entanglement contained in the state is persistent under particle loss [12,34].

Preparing collective dark states.—Let us now derive two quantum information theoretical schemes to prepare the state $|\psi_d^3\rangle$ deterministically. Below, we will present dissipative schemes to prepare them probabilistically. They can be generalized to preparing $|\psi_d^N\rangle$ for $N > 3$.

In both methods we initially prepare the state $|\psi_-\rangle = (|01\rangle - |10\rangle)/\sqrt{2}$ for two of the particles denoted as particles 1 and 2, which can be achieved by applying a CNOT to the two particles in the initial product state $(|0\rangle - |1\rangle)/\sqrt{2} \otimes |1\rangle$.

In the first method we then prepare particle 3 in the state $|2\rangle$ and apply the 3-qutrit gate $e^{-i2\pi/9(X\otimes X\otimes X+\text{H.c.})}$, where $X = |1\rangle\langle 0| + |2\rangle\langle 1| + |0\rangle\langle 2|$, in order to obtain the state $|\psi_d^3\rangle$ up to local phase gates. This preparation procedure can be verified easily realizing that $X^3 = \mathbb{1}$.

Alternatively, we prepare particle 3 in $|+\rangle = (|0\rangle + |1\rangle + |2\rangle)/\sqrt{3}$ and apply the two-qutrit unitary $U = |0\rangle\langle 0| \otimes X + |1\rangle\langle 1| \otimes X^2 + |2\rangle\langle 2| \otimes \mathbb{1}$ on the particle pairs (3,1) and (3,2) in order to obtain $|\psi_d^3\rangle$.

Let us point out, that given $|\psi_d^{N-1}\rangle$ for $N-1$ particles, the state $|\psi_d^N\rangle$ can be obtained by preparing particle N in $1/\sqrt{N}\sum_{i=0}^{N-1}(-1)^{(N-1)(1+i)}|i\rangle$ and applying $U = \sum_{i=0}^{N-1}|i\rangle\langle i| \otimes X^{i+1}$, where $X = |0\rangle\langle N-1| + \sum_{i=0}^{N-2}|i+1\rangle\langle i|$, to all particle pairs (N, j). Hence, $|\psi_d^N\rangle$ can be prepared recursively.

In a similar manner the state $|\psi_{\text{sr}}^N\rangle$ can be prepared by using $|\psi_+\rangle$ instead of $|\psi_-\rangle$ as the initial state of particles 1 and 2 and omitting the minus sign in the initial state of particle N. However, the properties of $|\psi_{\text{sr}}^N\rangle$ are very different from $|\psi_d^N\rangle$, as, e.g., $|\psi_{\text{sr}}^N\rangle$ has much less symmetries. Another difference can be found in the geometric measure of entanglement $E_g(|\psi_{\text{sr}}^N\rangle) = 1 - N!/N^N$ [33,35], which is much smaller than E_g for the dark state.

Dissipative generation of dark states.—As collective excitation and emission is inherently nonlocal and built into our model automatically, it will not only perturb a perfect dark state but one can achieve preparation via collective decay. In the following we will exhibit such collective dynamics of the system for various configurations.

Again, we consider the case of three atoms with two decay channels, i.e., three Λ systems, in a equilateral triangle or, alternatively, an equidistant chain and numerically solve for the dynamics.

A simple method to prepare the dark state probabilistically works as follows. We use the surprising fact that a nearby atom in the final state of a chosen transition can be utilized to suppress a particular decay channel of an atomic excitation. To see that, we start from an antisymmetric two-atom state $|\psi_d^2\rangle = (|g_1, e\rangle - |e, g_1\rangle)/\sqrt{2}$, which will not decay on the first transition to $|g_1\rangle$. This state, however, decays on the second transition towards $(|g_1, g_2\rangle - |g_2, g_1\rangle)/\sqrt{2}$. Now, let us add a third atom in either of the two ground states.

As shown in Fig. 3, a third atom prepared in $|g_1\rangle$ will not prevent decay (dotted red line), while a third atom in the

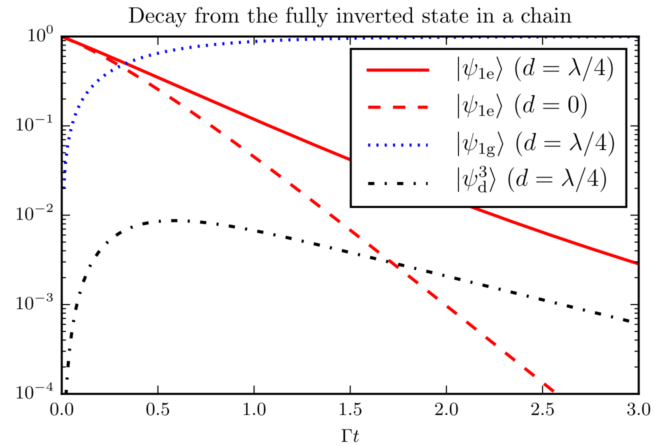

FIG. 4. Decay of three Λ-type atoms in an equidistant chain of distance $d = \lambda/4$ with nonequal Γ_j^{ik} starting from the totally inverted state $|eee\rangle$. The solid red line gives the excited state population per atom, the dotted blue line shows the population in the ground state subspace and the dashed-dotted black line gives the dark state $|\psi_d^3\rangle$ fraction during the decay. For comparison the red dashed line exhibits ideal collective decay at $d = 0$ with all equal Γ^{ik_j} (Dicke case). Note that during the evolution the dark (gray) state, which decays much slower, becomes populated partially. Again, we neglect Ω_j^{ik}, which would lead to an oscillatory behavior in the populations.

state $|g_2\rangle$ partially prevents decay and results in a finite excited state population probability at long times (dashed green line). Hence, after some time the system has either decayed to $(|g_1, g_2, g_2\rangle - |g_2, g_1, g_2\rangle)/\sqrt{2}$ or ends up in the dark state $|\psi_d^3\rangle$. Thus, preparing two atom dark states in the presence of other unpolarized ground state atoms is a key route for a probabilistic preparation of dark states. In this sense optical pumping as used in some experiments is counterproductive.

It is known that in spatially extended systems with nonuniform radiative coupling coefficients Γ_j^{ik} no perfect dark state but only long-lived subradiant states exist [36]. This implies that free space spontaneous decay from a multiply excited state can also sometimes end up in such a gray state [18] in close analogy to tailored deterministic entanglement generation between the ground states of interacting Λ atoms [37,38].

A central question now concerns the extent to which such a dissipative preparation works for several independent decay channels. By construction, the ideal dark state $|\psi_d^N\rangle$ is also decoupled from further symmetric laser excitation. Hence, the state is dark in absorption as well, similar to coherent population trapping in the ground state manifold [27].

For another conceptually simple approach to preparing the dark state we start from a totally inverted state $|eee\rangle$ for three atoms placed at a suitable finite distance, where the off diagonal elements of the matrix Γ_j^{ik} acquire negative values. As the dark component decays the slowest it should survive at the end. In Fig. 4 we demonstrate this mechanism for a three qutrit chain with a distance of $d = \lambda/4$. A comparison of the excited state fraction population for a finite sized chain (red line) with the ideal collective decay (dashed red line) shows a slowdown of the decay at late times, where indeed a small fraction of the population ends up in the dark state (black line). As the dark state has only little overlap with any product state, this fraction is small but can become relatively important at late times. Since $|\psi_d^3\rangle$ acquires a finite lifetime for finite distances, this fraction eventually decays as well but at a much slower rate.

Conclusions.—As our key result we show that the concept of dark or subradiant states can be generalized to multiple decay channels, if one includes one more particle than decay channels. The corresponding dark states are completely antisymmetric, highly entangled multipartite states with a plethora of quantum information applications. They can be prepared by a sequence of bipartite or tripartite gates or via tailored spontaneous emission from multiply excited states in optical lattices. A generalization to multiple excitations and several excited states as well as including the motional atomic degrees of freedom can be envisaged.

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