Quantum Engineering of Superdark Excited States in Arrays of Atoms

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We suggest a regular method of achieving an extremely long lifetime of a collective singly excited state in a generic small-size ensemble of $N$ identical atoms. The decay rate $\Gamma_N$ of such a ‘superdark’ state can be as small as $\Gamma_N \propto (\Gamma r/\lambda)^{2(N-1)}$ ($\Gamma$ is the radiative decay rate of an individual atom, $r$ and $\lambda$ are the system size and the wavelength of the radiation, respectively), i.e., considerably smaller than in any of the systems suggested up to now. The method is based on a special fine tuning of the atomic Hamiltonian: namely, on a proper position-dependent adjustment of atomic transition frequencies. So chosen set of the control parameters is sufficient to ensure the minimum of the spontaneous decay rate of the engineered state in a generic ensemble of atoms (‘qubits’).

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The phenomenon of subradiance (and superradiance as well) is one of the central points when discussing emission properties of systems of identical closely spaced atoms (qubits). There is growing interest in sub-wavelength atomic ensembles with an enhanced radiative lifetime of collective excitations. Slowly decaying (dark) states can be implemented for the storage of information in quantum memory devices.

Several configurations of one- and two-dimensional (1D and 2D) atomic arrays have been proposed (see, e.g. Ref. 11) that show substantial decrease of the decay rate as compared to that of an individual atom. For two atoms, a few schemes for controlling subradiant states in 1D 12–14 and in 3D 11,15 were suggested. An interesting (although rather complicated) scheme for constructing a singly excited subradiant state in a one-dimensional array of many atoms was considered in Refs. 14,16. As for the experiment, some evidence in favour of a change in the spontaneous decay rate was obtained, for example, in Ref. 16 for an ensemble of many atoms and in Ref. 17 for a system of two ions in a trap. Finally, subradiance from a cloud of cold atoms was reliably observed 15,19.

The suppression of the radiative decay is caused by destructive interference of emission amplitudes of different members of the atomic ensemble. This effect is most pronounced in the seminal Dicke model 20 of a compact ensemble of $N$ identical two-level atoms without non-retarded dipole-dipole interaction: one of the singly excited collective states is super-radiative, while the other $N-1$ singly excited states are non-radiative at all.

The presence of the dipole-dipole interaction leads to formation of the eigenstates (excitons) of the atomic ensemble which may reveal the properties of super- and sub-radiation. In small-size ensembles the resonant dipole-dipole excitation transfer is the strongest effect, while the spontaneous radiative decay occurs due to a weaker interaction with the transverse quantum electromagnetic field. Both the exciton states and their radiative decay rates are determined by the geometry of the atomic array.

The search for an array geometry with a minimal decay rate seems to be the matter of art, intelligence, and luck.

In this Letter we suggest a regular method to achieve an extremely long lifetime of a collective state in a generic small-size array of $N$ atoms. The decay rate $\Gamma_N$ of such a ‘superdark’ state can be as small as $\Gamma_N \propto (\Gamma r/\lambda)^{2(N-1)}$ ($\Gamma$ is the radiative decay rate of an individual atom, $r$ and $\lambda$ are, correspondingly, the system size and the resonant wavelength of the atomic transition). The method is based on a special fine tuning of the exciton Hamiltonian. We show that it is sufficient to adjust the frequencies of individual atomic transitions. We begin from an analysis of a generic atomic ensemble and after that we concentrate on the case of a finite regular chain of atoms.

We consider an ensemble of $N$ identical atoms located at spatial points $\mathbf{R}_j$ $(j = 1, 2, ..., N)$. Each of singly excited collective atomic states $|\mathbf{C}\rangle$ can be represented as a superposition $|\mathbf{C}\rangle = \sum_{j=1}^{N} C_j |\mathbf{e}_j\rangle$ of basis states $|\mathbf{e}_j\rangle$ where the $j$-th atom is in its excited state $|\mathbf{e}_j\rangle$ while all the other atoms are in the ground state $|\mathbf{g}\rangle$. These exciton states are eigenstates of the atomic ensemble Hamiltonian $\hat{H}^{(at)}$: $\hat{H}^{(at)}|\mathbf{C}\rangle = E_{\mathbf{C}}|\mathbf{C}\rangle$, or in the matrix form (we put $\hbar = 1$)

$$\omega_j C_j + \sum_{j' = 1}^{N} U(\mathbf{R}_j, \mathbf{R}_{j'}) C_{j'} = E_{\mathbf{C}} C_j .$$  \hspace{1cm} (1)

Here the prime at the sum symbol means the exclusion of the term $j' = j$; $\omega_j$ is the transition frequency of the $j$-th atom; $U(\mathbf{R}_j, \mathbf{R}_{j'})$ is the matrix element of the dipole-dipole interaction between the states $|\mathbf{e}_j\rangle$ and $|\mathbf{e}_{j'}\rangle$; its particular form will be specified below. For the introductory illustration of the suggested approach we assume that the excited atomic state is non-degenerate. Then we will describe a more general situation. The radiative decay rate of a given exciton state is determined by the
Fermi Golden Rule:

$$\Gamma_N \propto \Gamma \sum_{\nu=1,2} \left| \sum_{j=1}^N (d \cdot e^{(\nu)}_{j}) e^{i k R_j C_j} \right|^2 = \Gamma \sum_{j,j'=1}^N C_j W_{j,j'} C_{j'}, \quad (2)$$

where \(d = (e|d|g)\) is the matrix element of the dipole moment operator and \(e^{(\nu)}_{j}\) is the unit polarization vector of a photon of the wave vector \(k\). The integration in Eq. (2) is performed over the solid angle in the momentum space where \(k = |k| = 2\pi/\lambda\) is fixed by the energy of the state \(|C\rangle\). The first sum runs over the polarizations. The quadratic form in Eq. (2) is positively defined. The matrix \(W_{j,j'}\) is obviously Hermitian. Moreover, it is real due to the symmetry of the integrand in Eq. (2) with respect to the change \(k \rightarrow -k\). Therefore, for a given arrangement of atoms (i.e., for a given matrix \(W_{j,j'}\)), there exists a real unit vector \(\{C_j\} (j = 1, \ldots, N; \sum_j C_j^2 = 1)\) that provides the minimal value of the quadratic form and, therefore, realizes the minimal decay rate. If the corresponding state \(|C\rangle = \sum_{j=1}^N C_j |j\rangle\) were an eigenstate of the atomic Hamiltonian \(\hat{H}(\alpha)\), the problem of finding the state with the smallest decay rate would be solved. However, in general, the two matrices, \(W_{j,j'}\) and \(\hat{H}(\alpha)\), do not have a common eigenvector. Our current task is to perform a fine tuning of the atomic Hamiltonian in order to make the state \(|C\rangle\) its eigenstate obeying the system of equations (1). In these equations the matrix \(U(R_j, R_{j'})\) of the dipole-dipole interactions is fixed by the positions of the atoms, while the local frequencies \(\omega_j\) can be controlled by external fields. It is naturally to count these shifts (and the eigenenergy \(E\) as well) from an ensemble averaged frequency \(\overline{\omega}_0\), so that \(\sum_{j=1}^N \omega_j = 0\). This condition together with \(N\) equations (1) with fixed \(\{C_j\}\) uniquely determine \(N+1\) real quantities: local frequency shifts \(\omega_j\) and the eigenenergy \(E\), as

$$E = \frac{1}{N} \sum_{j,j'=1}^N \frac{1}{C_j} U(R_j, R_{j'}) C_{j'}, \quad (3)$$

$$\omega_j = E - \frac{1}{C_j} \sum_{j'=1}^N U(R_j, R_{j'}) C_{j'}. \quad (4)$$

This solves the problem of the Hamiltonian tuning to ensure the existence of an eigenstate with the minimal decay rate for the given geometry of the atomic ensemble.

The considered model can be generalized to the case where the non-degenerate ground atomic state has the angular momentum \(J = 0\) while the excited state is degenerate having the angular momentum \(J = 1\) with projections \(m = 0, \pm 1\) on the quantization axis \(\hat{z}\). Hereafter we shall use an equivalent basis of atomic excited states \(|\alpha\rangle (\alpha = x, y, z)\), connected with the states \(|m\rangle\) by \(|m = 0\rangle = |z\rangle, |m = \pm 1\rangle = (|x\rangle \pm i|y\rangle)/\sqrt{2}\). Matrix elements of the Cartesian components \(d_\beta (\beta = x, y, z)\) of the dipole moment operator in this representation are \(d_\beta \langle g | \delta \rangle = d_\delta \delta_{\beta, \delta}\). The operator of the dipole-dipole interaction between the atoms is

$$\hat{H}_d = \sum_{j,j'=1}^N \frac{\langle \mathbf{d}_j \cdot \mathbf{d}_{j'} \rangle - 3(\mathbf{d}_j \cdot \mathbf{n}_{j,j'})(\mathbf{d}_{j'} \cdot \mathbf{n}_{j,j'})}{|R_j - R_{j'}|^3}, \quad (5)$$

where \(\mathbf{d}_j\) is the dipole operator of the \(j\)-th atom; the unit vector \(\mathbf{n}_{j,j'} = (R_j - R_{j'})/|R_j - R_{j'}|\). In general, the operator \(\hat{H}_d\) mixes excited states with different projections \(\alpha\). Therefore, collective singly excited eigenstates of the atomic array are described by a superposition

$$|C\rangle = \sum_{\alpha=1}^N \sum_{j=1}^N C_j^{(\alpha)} |\alpha; j\rangle, \quad \sum_{\alpha=1}^N \sum_{j=1}^N |C_j^{(\alpha)}|^2 = 1. \quad (6)$$

Here the sum runs over \(\alpha = x, y, z\); the basis vector \(|\alpha; j\rangle\) denotes the state where the \(j\)-th atom is in its excited state \(|\alpha\rangle (J = 1)\) while all the other atoms are in their ground states \((J = 0)\). Correspondingly, the equations for the eigenstates and for the radiative decay rate look similar to Eqs. (1) and (2) but with the matrices \(U_{\alpha,\beta}(R_j, R_{j'})\) and \(W_{\alpha,\beta}^{(\alpha)}\) having the additional indices. Interaction of the \(j\)-th atom with the plane wave of wave vector \(\mathbf{k}\) and polarization \(\nu\) is governed by the operator \(H_{int} \propto - (\mathbf{d} \cdot e^{(\nu)}_{j}) e^{i k R_j}\). The spontaneous decay rate of the state \(|C\rangle\) is given by

$$\Gamma_N = \frac{3 \Gamma}{8\pi} \int_0^{2\pi} d\varphi \int_0^\pi d\theta \frac{\pi}{2} \sum_{\nu=1}^2 \left| \sum_{\alpha=1}^N \sum_{j=1}^N e^{(\nu)}_{j,\alpha} C_j^{(\alpha)} e^{i k R_j} \right|^2 = \Gamma \sum_{\alpha,\beta=1}^N C^{(\alpha)}_j W^{\alpha,\beta}_{j,j'} C^{(\beta)}_{j'}. \quad (7)$$

where the wave vector is parameterized as \(\mathbf{k} = k (\sin \varphi \cos \varphi, \sin \varphi \sin \varphi, \cos \varphi)\). The polarization vectors are chosen as \(e^{(1)}_{j,\alpha} = (-\sin \varphi, \cos \varphi, 0)\) and \(e^{(2)}_{j,\alpha} = (\cos \varphi \cos \varphi, \cos \varphi \sin \varphi, -\sin \varphi)\); their Cartesian components obey the relation \(\sum_{\nu=1}^2 e^{(\nu)}_{j,\alpha} e^{(\nu')}_{j',\beta} = \delta_{\alpha,\beta} - k_\alpha k_\beta/k^2\).

Using Eq. (7) one can calculate the decay rate of any singly excited eigenstate of the atomic Hamiltonian. The matrix \(W^{\alpha,\beta}_{j,j'}\) is Hermitian, and real due to the symmetry \(\mathbf{k} \rightarrow -\mathbf{k}\). The matrix \(U^{\alpha,\beta}(R_j, R_{j'})\) corresponding to the matrix elements of the Hamiltonian \(\hat{H}_d\) between the states \(|\alpha\rangle\) is obviously real, too. Thus, similarly to the simplified model, we can find the real vector \(|C\rangle\) that realizes the minimum of the quadratic form \(\Gamma_N\). Then we can adjust the local atomic transition frequencies making this optimal vector to be an eigenvector of the atomic Hamiltonian. In this way, the conceptual problem of the proper tuning of the atomic Hamiltonian to ensure the existence of an eigenstate with the minimal (for the given array geometry) decay rate is solved.
For atomic ensembles of a low symmetry (where excitation states are not characterized by a single atomic polarization $\alpha$) implementation of the described procedure may be complicated because it would require independent tuning of all three initially degenerate atomic transition frequencies of each atom. The situation simplifies for geometries where the dipole-dipole interaction does not mix excited states of the atomic Hamiltonian with different polarizations $\alpha$. This happens in configurations where the vectors connecting atom sites are parallel or perpendicular to the direction of the transition dipole moment of a chosen excitation polarization. Such are, for instance, a plane ensemble and a linear chain of atoms.

Now, we study a linear chain of atoms located at the sites $\mathbf{R}_j = R_j \hat{z}$. We will explicitly construct the ‘optimal’ vector \(|\mathbf{C}\rangle\) and the atomic Hamiltonian so that the decay rate of the found ‘superdark’ state will be enormously small $\Gamma_N = \Gamma(r/\lambda)^{2(N-1)}$. For the excitation polarized parallel (perpendicular) to the chain directions, i.e., for $|\alpha\rangle = |z\rangle$ ($|\alpha\rangle = |x\rangle$) or $|\alpha\rangle = |y\rangle$), matrix elements of the operator \(\mathbf{R}_j\) for the excitation transfer between the sites $R_j$ and $R_{j'}$ are given by

$$U_{j,j'} = -\frac{\partial^2}{\partial R_j^2} \quad \text{and} \quad U_{j,j'}^+ = \frac{\partial^2}{\partial R_j^2},$$

where $R_{j,j'}$ is the distance between the sites $j$ and $j'$ of the chain. The expression \(\Gamma_N\) for the decay rate of the collective singly excited state of the polarization $\alpha = z$ (\(|\rangle\)), or $\alpha = x, y$ (\(\perp\)), reduces to

$$\Gamma_N = \frac{3\Gamma}{8\pi} \int d\omega f^{(\alpha)}(\omega, \theta, \varphi) \left| \sum_{j=1}^N C_j^{(\alpha)} e^{ikR_j \cos \theta} \right|^2,$$

where $f^{(\alpha)}(\omega, \theta, \varphi) = 1 - k_{\alpha}^2/\omega^2$, i.e., $f^{(z)}(\theta, \varphi) = \sin^2 \theta$ and $f^{(x)}(\theta, \varphi) = 1 - \sin^2 \theta \cos^2 \varphi$. The integration determines the corresponding matrix $W_j^{\alpha,\alpha'}$ of the quadratic form \(\mathbf{W}\):

$$W_{j,j'}^{\alpha,\alpha'} \equiv W_{j,j'}^{(\alpha,\alpha') \parallel} = 3 \frac{\sin \xi - \xi \cos \xi}{\xi^3};$$

$$W_{j,j'}^{\alpha,\alpha'} \equiv W_{j,j'}^{(\alpha,\alpha') \perp} = \frac{3}{2} \frac{\xi \cos \xi + (\xi^2 - 1) \sin \xi}{\xi^3},$$

where the notation $\xi = kR_{j,j'}$ is used for brevity. Exact eigenvectors corresponding to the minimal eigenvalues of these matrices can be determined numerically. To make a simple analytical estimation of the minimal decay rate for a short chain (of the length $r \ll \lambda$), it is natural to expand quantities in $\sqrt{kr}$ in powers of the small quantities $kR_j \cos \theta$:

$$\left| \sum_{j=1}^N C_j^{(\alpha)} e^{ikR_j \cos \theta} \right| = \sum_{n=0}^\infty \frac{(ikr \cos \theta)^n}{n!} \sum_{j=1}^N \left( \frac{R_j}{r} \right)^n C_j^{(\alpha)}.$$  

To minimise this expression (therefore, $\Gamma_N$) we require vanishing of $N-1$ first terms ($n = 0, \ldots, N-2$) of the expansion in $(kr)^n$. This means imposing $N-1$ (i.e., the maximal possible number) linear constraints on the $N$-component unit vector \(|\mathbf{C}\rangle\):

$$\sum_{j=1}^N \left( \frac{R_j}{r} \right)^n C_j^{(\alpha)} = 0; \quad n = 0, 1, \ldots, N-2. \quad (13)$$

Under conditions \(13\) the expansion \(12\) begins with terms $\propto (kr)^{N-1}$ and we arrive at the announced estimate for the minimal decay rate $\Gamma_N \propto (kr)^{2(N-1)}$. This minimal value realized at the optimal vector determined by \(13\), is very fragile: a slightest deviation of the state \(|\mathbf{C}\rangle\) from the optimal one would involve terms of the expansion \(12\) with lower powers of $kr$ (i.e., of a much larger value). This results in a strong sharpness of the decay rate dependences on tuning parameters (see below Fig. 1 and Fig. 2 for particular examples).

A formal solution to the system \(13\) can be expressed in terms of the $N \times N$ Vandermonde matrix $\mathbf{V}$ ($V_{ij} = (R_i/r)^j$; $i = 0, \ldots, N-1$; $j = 1, \ldots, N$): \(C_j^{(\alpha)} = c(\mathbf{V}^{-1})_{\alpha,j}, N\), where $c$ is determined by the normalization condition.

Explicit results can be presented for a chain of equally spaced atoms $R_j = (j-1)a$; $j = 1, \ldots, N$. In this case the exact (normalized) solution to \(13\) is expressed in terms of the binomial coefficients:

$$C_j^{(\alpha)} = (-1)^{j-1} \frac{[(N-1)!]^2}{(j-1)!(N-j)! \sqrt{(2N-2)!}}.$$

Note that this solution is either symmetric (for odd $N$) or antisymmetric (for even $N$) with respect to the replacement $j \rightarrow N-j+1$:

$$C_j^{(\alpha)} = (-1)^{N-1} C_{N-j+1}^{(\alpha)}.$$  

The decay rate \(\Gamma_N\) of this optimal state \(|\mathbf{C}\rangle\) is

$$\Gamma_N = \frac{3\Gamma}{4N-2} \frac{[(N-1)!]^2}{(2N-2)!} (ka)^{2(N-1)} \left\{ \begin{array}{c} 1 \\ N \end{array} \right\}$$

where the symbol $\parallel$ (\(\perp\)) denotes the excitation polarization parallel (perpendicular) to the chain. Now, to make the found optimal state \(|\mathbf{C}\rangle\) an eigenstate of the atomic Hamiltonian, we perform the fine tuning of the local atomic frequencies. The adjusted atomic frequencies are given by Eq. \(\ref{eq:freq}\), where the matrix of the dipole-dipole interaction for the considered geometry takes the form (see Eq. \(\ref{eq:mat}\)): \(U^{\parallel}(\mathbf{R}_j, \mathbf{R}_j') = -2d(k/j^3)\parallel \) or \(U^{\perp}(\mathbf{R}_j, \mathbf{R}_j') = d(k/j^3)\parallel\). The eigenenergy $E$ of the constructed state \(|\mathbf{C}\rangle\) is given by Eq. \(\ref{eq:energy}\). As a consequence of Eq. \(\ref{eq:energy}\), the shifted atomic frequencies possess the symmetry \(\omega_j = \omega_{N-j+1}\). The both decay rates \(\ref{eq:gamma}\) are very small when the system size is small as compared to the resonance wavelength, $(N-1)a \ll \lambda$. The same condition is sufficient to treat the decay rate \(\ref{eq:gamma}\) of the state \(\ref{eq:state}\) independently of other eigenstates of the atomic ensemble.
What is exciting—the property of ‘superdarkness’ is extremely sensitive to the shift(s) of transition frequencies [21]. This is illustrated by the results of numerical calculations for the chain of three and four equally spaced atoms. For three atoms, the only adjusted parameter is the shift of the transition frequency of the middle atom with respect to those of the edge atoms: Ω = ω2 − ω1. To characterize the subradiance dependence on this shift we introduce the quantity Γ(Ω) that is the slowest decay rate of those for all eigenstates. The dependences of so defined Γ on Ω/U are shown in Fig. 1 for two values of the parameter (ka)2; here U is the dipole-dipole interaction between the neighboring atoms: U⊥ = d2/a3 and U∥ = −2d2/a3 [see Eq. (3)]. Notice that the sign of the optimal shift is negative for the perpendicular polarization (Γ⊥) and positive for the parallel polarization (Γ∥). One can see very sharp minima for all curves, their abscessas being very close to the ‘optimum asymptotic value’ Ω/Ur defined by Eq. (11) with the eigenvector [14]. For N = 3, Ω/Ur = −7/8. Meanwhile, the optimum is actually reached at Ω/Ur that is a little shifted as can be seen in Fig. 1. At, e.g., ka = 0.1, the mismatch is ΔΩ/Ur ≈ 3.0 · 10−3 for and, in a good approximation, scales ∝ (ka)2. At the same time, the corresponding decay rates lie (due to the extreme sharpness) a few lower than those defined by asymptotic Eq. (11). E.g., the fall is ∝ 2.64 times for ΓN=3 at ka = 0.1.

Similar results for four atoms are given in Fig. 2. There is also only one adjusted parameter, i.e. the transition frequency shift Ω for the two middle atoms with respect to the edge atoms. In the case of N ≥ 5 atoms the [(N − 1)/2] parameters must be adjusted. We don’t draw the corresponding multidimensional pictures—only note that the minimum near the set of optimum values defined by Eq. (4) with the eigenvector [14] is even sharper than in the just considered cases N = 3 and N = 4 (Figs. 1 and 2, respectively).

How strong could be the suppression of the decay rate Γ is shown in Table 1, where the values of ΓN for the optimally adjusted atomic frequencies are compared with those in the case where are no shifts, i.e. the transition frequencies of all atoms are equal. The same several orders of magnitude are in play comparing with the results for earlier treated subradiative systems including a very recently suggested ensemble in the form of a regular polygon [25] where ΓN ∝ Γ(π/λ)2N/[2]. The two exceptions are: (i) two separated by the distance nλ/2 atoms in 1D [3, 9]; (ii) extension of this idea to 3D using two similarly separated two-dimensional arrays of atoms [24]. However, the chain configuration is much more commonly considered due to its implementation using the optical
lattices, even subwavelength ones [23, 24]. Our examples above relate just to such a scheme at $ka \leq 1$ (for matching with $ka > 1$ see Ref. [27]). Certainly, usage of different tweezers for different atoms can be assumed. In addition we may suggest, in view of the both $N = 3$ and $N = 4$ cases, a speculative scheme of laser control using the standing wave as shown in Fig. 3.

To conclude, in this Letter the recipe is presented how to achieve a huge gain in subradiance of atomic ensembles. This gain is reached in a regular way by adjustment of atomic transition frequencies $\omega_j$. The demonstrated extreme sensitivity of the slow spontaneous decay rate and the accompanying narrow radiative width to external fields may be useful for precision measurements and diagnostics in addition to its undoubted fundamental significance.

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FIG. 3. (color online) Scheme for inducing an energy shift of the ground state in the middle atom(s). The nodes are at the positions of the edge atoms. The laser frequency is detuned relative to the frequency of the transition from the ground state to some high-lying excited state $|e\rangle$ of the atom(s). As shown in the diagram, this shifts the frequency of the transition $|e\rangle \rightarrow |g\rangle$ to the red, i.e. suitably for the perpendicular polarization. For the case of the parallel polarization the sign of the detuning should be reversed.
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