Single Photon Subradiance: Quantum Control of Spontaneous Emission and Ultrafast Readout

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Single Photon Subradiance:

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Recent work has shown that collective single photon emission from an ensemble of resonate two-level atoms, i.e. single photon superradiance, is a rich field of study. The present paper addresses the flip side of superradiance, i.e. subradiance. Single photon subradiant states are potentially stable against collective spontaneous emission and can have ultrafast readout. In particular it is shown how many atom collective effects provide a new way to control spontaneous emission by preparing and switching between subradiant and superradiant states.

Group theory is one of the most beautiful subjects in physical mathematics. No better example than Dicke superradiance [1–5]. Dicke taught us that radiating two level atoms can be insightfully grouped into angular momentum multiplets. To motivate this connection we note that each two level atom is a spinor. Then for two atoms (or two neutrons in a magnetic field, etc.) it takes four states to cover the spin space. The four spin states can be grouped into the spin triplet and singlet states depicted in the upper right corner of Fig. 1. As an example of the utility of the method the decay states of the system can now be read off using angular momentum matrix elements. Indeed the term "superradiance" derives from the fact that the decay rate from state $|r, m\rangle$ to $|r, m-1\rangle$ goes like the square of the angular momentum lowering operator connecting these states which is given by $(r+m)(r-m+1)$. Hence when $m = 0$ (equal population in the ground and excited states) the radiation emission rate goes as N^2. This superradiant rate can be understood semiclassically by noting that the electric field emitted by N coherently prepared dipoles is proportioned to N, and the intensity goes as N^2. In fact most of the calculations associated with superradiance experiments are carried out using a semiclassical Maxwell-Bloch formalism.

However, recent work has shown that collective single photon spontaneous emission from an ensemble of resonate two-level atoms is a rich field of study [6–11]. For example single photon superradiance from an ensemble much larger than the radiation wavelength yields enhanced directional spontaneous emission; and when the ensemble becomes larger than the radiation wave packet [12] interesting collective superradiant effects abound.

As is stated in the abstract and discussed below following, Eq. (2), the present focus is on control of spontaneous emission and the switching from subradiant to superradiant states. Application to single photon devices is apparent, but the study of cooperative subradiance is of interest in and of itself.

The physics of the subradiant states was explained by Dicke [1]. These states are necessary to span the N atom space. For example, the subradiant states are used in calculating the many atom Lamb shift via the timed Dicke states [12], but they have not stimulated anything like the amount of work that the superradiant state has. to be sure, interesting work on Dicke subradiance has been reported. For example, Pavolini and coworkers [13] observed a reduced emission rate due to "trapping" of a fraction of the atoms in a mixture of many subradiant states following pulsed excitation. Some experiments of interest have focused on preparation of the subradiant state of a two atom system [14]. Other interesting

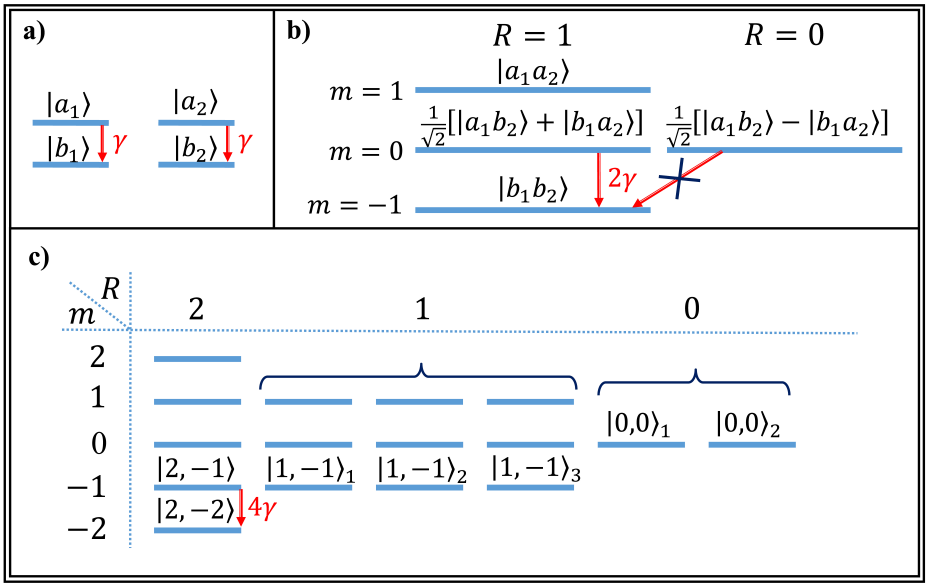

Fig. 1: The 16 states from a four-atom system are depicted. a) The upper (lower) states for atoms $j = 1, 2$ are $|a_j\rangle$ ($|b_j\rangle$). b) The two atoms can be grouped into triplet $R = 1$ and singlet $R = 0$ state, where $R = N/2$ is the cooperation number, and m is $(N_a - N_b)/2$ where N_a and N_b are the number of atoms in $|a\rangle$ and $|b\rangle$. c) The group characterization of the 16 atom states is depicted in an $|R, m\rangle_p$ notation where the p index labels the column within a given R. The states $R = 1$ and $R = 0$ are 3 and 2 fold degenerate. The superradiant state $|2, -1\rangle$ decays at a rate four times that of a single atom. The states $|1, -1\rangle_s$ ($s = 1, 2, 3$) contain one photon energy and do not decay. It may be noted for $R = 1$ the s index denotes the number of singlet states, see e.g. Table I. The states $|0, 0\rangle_1$ and $|0, 0\rangle_2$ are two photon subradiant states involving 1 and 2 pairs of singlet states.

work [15] on an N atom inhomogeneously broadened ensemble driven by a weak pulse has demonstrated about 0.1% in a mixture of subradiant states. Subradiance in molecular [16] and quantum dot [17] systems is also of interest. We here present and analyze a simple method whereby a substantial fraction, in principle 100%, of the atoms can be placed in the single N atom subradiant state given by Eq. (2). Furthermore, it is possible to switch between the subradiant state of Eq. (2) to the companion superradiant state of Eq. (1) by 2π cycling of half the atoms as depicted in Fig. 2.

For our purposes it takes at least four atoms to introduce the story. The 2^4 states of the four spin system of Fig. 1 and Table 1 are spanned by the angular momentum multiplets having total "angular momentum" $R = 2$, 1 and 0.

Subradiant State	Singlet State Representation
$\|1,-1\rangle_1$	$\|s_{12}\rangle\|b_3b_4\rangle$
$\|1,-1\rangle_2$	$\big[\|s_{13}\rangle\|b_2b_4\rangle + \|s_{23}\rangle\|b_1b_4\rangle\big]/\sqrt{3}$
$\|1,-1\rangle_3$	$\big[\|s_{14}\rangle\|b_2b_3\rangle + \|s_{24}\rangle\|b_1b_3\rangle + \|s_{34}\rangle\|b_1b_2\rangle\big]/\sqrt{6}$
$\|0,0\rangle_1$	$\|s_{12}\rangle\|s_{34}\rangle$
$\|0,0\rangle_2$	$\big[\|s_{13}\rangle\|s_{24}\rangle + \|s_{23}\rangle\|s_{14}\rangle\big]/\sqrt{3}$

Table I: Subradiant states for a 4-atom system in terms of 2 atom singlet states, where $|s_{ij}\rangle = [|a_ib_j\rangle - |b_ia_j\rangle]/\sqrt{2}$. The notation is explained in Fig. 1. See also Supplement B.

As discussed in the preceeding, we seek single photon subradiant states which are long lived and can be switched to the single photon superradiant state given by

$$|+\rangle_N = \frac{1}{\sqrt{N}}\sum_{j=1}^{N}|j\rangle, \tag{1a}$$

where $|j\rangle = |b_1\cdots a_j\cdots b_N\rangle$ and $b_j(a_j)$ is the ground (excited) state of the jth atom. The companion subradiant state of Fig. 2b is given by

$$|-\rangle_N = \frac{1}{\sqrt{N}}\left[\sum_{j=1}^{N/2}|j\rangle \;\; - \sum_{j'=N/2+1}^{N}|j'\rangle\right]. \tag{1b}$$

In the following we first investigate single photon superradiant in small and extended samples using single photon preparation. And, as is shown in Supplement A, the $|+\rangle_N$ state decays at the superradiant state rate $N\gamma$, where γ is the single atom decay rate and the $|-\rangle_N$ state is long lived. In both cases post selection allows for a thin optical medium on preparation and a thick medium on decay. Subradiant state preparation of a four atom system to state $|0,0\rangle$ and to N atom super- and sub-radiant states Eq. (3a,b) without post selection is also presented. We conclude with a brief summary of results and open questions.

It is useful to write the $|-\rangle_N$ state in terms of the $|s_{jj'}\rangle$ singlet states of Table I, where the j index runs from 1 to $N/2$ and j' runs from $N/2+1$ to N; that is

$$|-\rangle_N = \frac{1}{\sqrt{N/2}}\sum_{j,j'}|s_{jj'}\rangle|\{b\}_{jj'}\rangle, \tag{2}$$

where $|\{b\}_{jj'}\rangle$ is the N atom ground state with the j and j' atoms removed. Having stored a photon in state $|-\rangle_N$ we can extract, i.e. readout this information by switching the minus

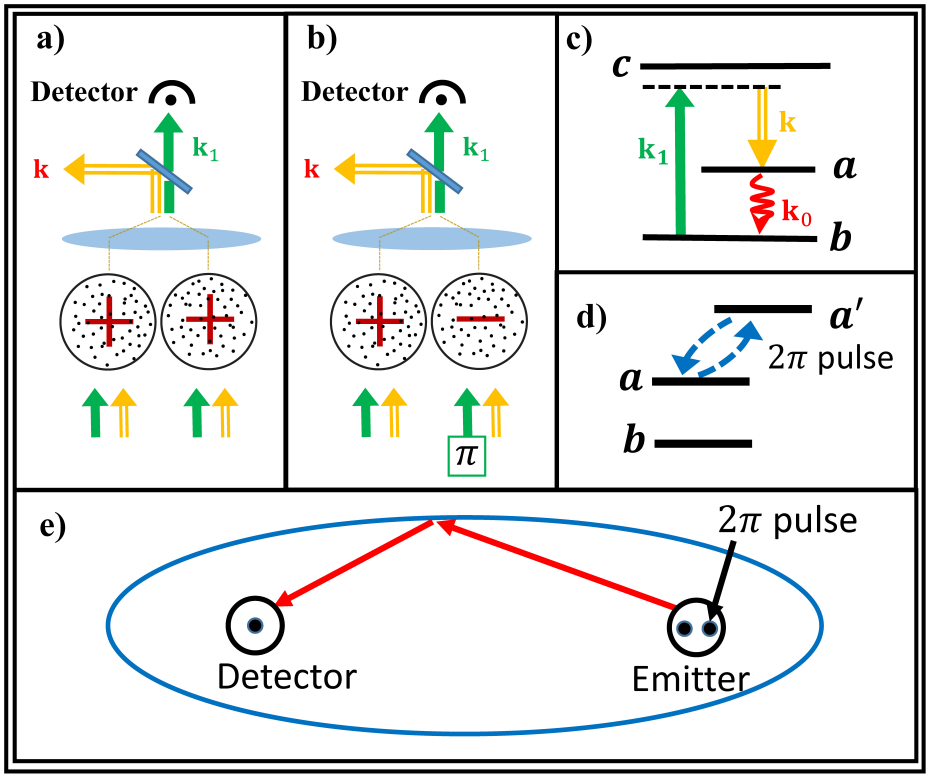

Fig. 2: a) Excitation of $|+\rangle_N$ state. A single photon of wave vector $\mathbf{k}_1$ is accompanied by a laser having wave vector $\mathbf{k}$, $\mathbf{k} - \mathbf{k}_1 = \mathbf{k}_0$. The $\mathbf{k}_0$ photon is resonant with the transition $|a\rangle$ to $|b\rangle$. The atoms are weakly driven by the excitation process i.e. the atomic medium is optically thin during the preparation process; the $\mathbf{k}$ radiation is isolated from the detector. The lack of a count heralds the preparation of the $|+\rangle_N$ state. b) Same as part (a) but single photon $\mathbf{k}$ is divided by a beam splitter and shifted by π on the RHS so those atoms are prepared out of phase with the LHS atoms. That is the $|a_{j'}\rangle$ atoms on the RHS are multiplied by -1; the net result in (a) and (b) is that a no-count event signals the fact that the $|\pm\rangle_N$ state has been prepared. c) The atoms are weakly driven by a Raman-type process in which the $\mathbf{k}_1$ photon excites the atom to a virtual state and the $\mathbf{k}$ photon takes the atom to the $|a\rangle$ state. d) Cycling the RHS atoms $a \to a' \to a$ results in another factor of -1, which when applied to the RHS atoms takes the single photon subradiant state $|-\rangle$ to $|+\rangle$. e) Sketch of double microdot emitter which is small compared to the superradiant wavelength ($\lambda_0 > 1\mu$). As in Fig. 2b the double dots are initially prepared in $|-\rangle_N$; and then after some storage time ($T >$ millisec) switched to the $|+\rangle_N$ state by the 2π pulse which drives the RHS dot as depicted in Fig. 2d. The ellipsoidal cavity directs all λ_0 photons emitted to the detector. The cavity walls are transparent to the preparation radiation $\mathbf{k}_1$ and $\mathbf{k}$ of Fig. 2c as well as the 2π switching pulses.

sign in Eq. (1b) to a plus. This we do by cycling the RHS j' atoms with a 2π pulse as in Fig. 2d. This results in $|a_{j'}\rangle$ going to $-|a_{j'}\rangle$ and we change the subradiant state Eq. (1b) to the symmetrical state $|+\rangle$ given by Eq. (1a), which decays at the rate $\Gamma_N = N\gamma$. Thus the single photon subradiant state $|-\rangle$ is stable against collective spontaneous emission with storage time long compared with Γ_N^{-1}. And can be addressed in ultrashort switching times of order $(N\gamma)^{-1}$, see Fig. 2e.

By way of comparison a photon stored in a high Q cavity has a lifetime of several μsec, and the cavity switching times as determined electronically are usually in the nanosecond range. Atomic dark states can also store a photon for long times but the switch out times are of order γ^{-1}.

These results have an extension to the large sample (timed Dicke) case. Then, as is shown in [6], the superradiant state prepared by a photon of vector $\mathbf{k}_0$ is given by

$$|+\rangle_{\mathbf{k}_0} = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{i\mathbf{k}_0\cdot\mathbf{r}_j} |b_1 b_2 \cdots a_j \cdots b_N\rangle, \tag{3a}$$

and as discussed in Supplement B the corresponding single photon subradiant state is given by

$$|-\rangle_{\mathbf{k}_0} = \frac{1}{\sqrt{N}_2} \sum_{j,j'}{}^{2} \frac{1}{\sqrt{2}} \left(|a_j b_{j'}\rangle e^{i\mathbf{k}_0\cdot\mathbf{r}_j} - |b_j a_{j'}\rangle e^{i\mathbf{k}_0\cdot\mathbf{r}_{j'}} \right) |\{b\}_{jj'}\rangle. \tag{3b}$$

Then, the $|+\rangle_{\mathbf{k}_0}$ state decays approximately at the rate

$$\Gamma_+ \cong \frac{3}{16\pi}\gamma\frac{\lambda^2}{A}(N-1) + \frac{\gamma}{2}, \tag{4a}$$

and the corresponding $|-\rangle_{\mathbf{k}_0}$ state decay rate goes as

$$\Gamma_- \cong \frac{3}{16\pi}\gamma\frac{\lambda^2}{A}\left(\frac{N}{2} - \frac{N}{2}\right) + \frac{\gamma}{2} - \frac{3}{16\pi}\gamma\frac{\lambda^2}{A}. \tag{4b}$$

It is important to note that Γ_- is lower bounded by the natural single atom rate γ. But the collective decay rates Γ_+ and Γ_- are very different and one can envision cases where a storage time of order γ^{-1} with directional photons emitted in times of order Γ_+^{-1} would be of interest. However in the interest of simplicity, the focus of the present paper is the preparation of the $|-\rangle_N$ state and subsequent switching between the single photon subradiant states, neighboring single photon superradiant and two photon subradiant states.

As an aside we note that higher energy states can be realized by driving the system with the symmetric raising operator $\hat{R}^+ = \sum_j \hat{\sigma}_j^+$. In particular the $|-\rangle_N$ state of Eq. (2) is promoted to the two photon state

$$|-\rangle_N^{(2)} = \frac{1}{\sqrt{N/2}} \sum_{j,j'} |s_{jj'}\rangle |+_{jj'}\rangle \tag{5}$$

where $|+_{jj'}\rangle$ is the symmetric state Eq. (1) with j and j' atoms missing. This example makes clear the utility of the writing the subradiant states in the $|s_{jj'}\rangle|\{b\}_{jj'}\rangle$ form of Eq. (2), namely the raising $\hat{R}^+$ operator only acts on the $|\{b\}_{jj'}\rangle$ states since $\hat{R}^+|s_{jj'}\rangle = 0$.

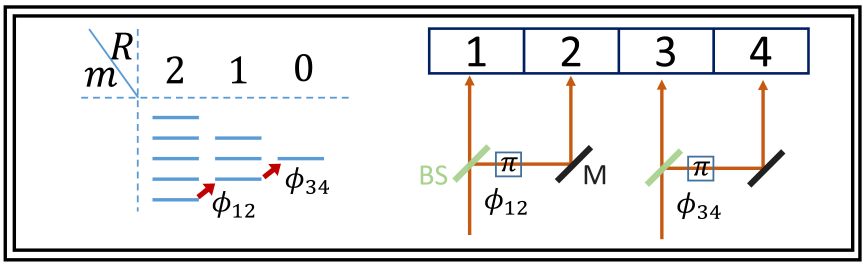

Fig. 3: The single photon subradiant state $|1,-1\rangle_1$ is prepared by coupling atoms 1 and 2 in $|2,-2\rangle = |b_1 b_2 b_3 b_3\rangle$ with the single photon state $|\Phi\rangle_{12}$ of Eq. (6) prepared via a π phase shift as indicated. The double photon subradiant $|0,0\rangle_1 = |s_{12}s_{34}\rangle$ is prepared by coupling atoms 3 and 4 in $|1,-1\rangle_1 = |s_{12}\rangle|b_3 b_4\rangle$ with $|\Phi\rangle_{34}$. BS and M denotes beam splitters and mirrors and $\boxed{\pi}$ indicates a π phase shifter.

Next we turn to the four atom case of Fig. 1 and consider preparation of single photon $|1,-1\rangle_1$ and two photon $|0,0\rangle_1$ subradiant states. Here we are not using weak Raman excitation and do not require post selection. Consider the four atom system in Fig. 3. There we see the four atom states $|1,-1\rangle_1$ and $|0,0\rangle_1$ prepared by sidewise illumination. In particular, the single photon subradiant state $|1,-1\rangle_1$ is prepared by passing a single photon $|\gamma\rangle$ through a beam splitter, and phase shifting the radiation directed to atom 2 by π as in Fig. 3. The light in the two legs of the optical path interacts with atoms 1 and 2 and post-selecting the photon vacuum then prepares the singlet state $|s_{12}\rangle|b_3 b_4\rangle$. That is, beginning with the photon state

$$|\Phi\rangle_{12} = \frac{1}{\sqrt{2}} \left(|1_1, 0_2\rangle - |0_1, 1_2\rangle\right), \tag{6}$$

where atoms 1 and 2 are driven by photons $|1_1\rangle$ and $|1_2\rangle$ via the resonant interaction

$$V = \frac{1}{2}\hbar g \sum_{i=1,2} (\hat{\sigma}_i^+ \hat{a}_i + \hat{a}_i^\dagger \hat{\sigma}_i)$$

where $\hbar g$ is the product of the atomic matrix element $\wp$ and the electric field per photon $\mathcal{E} = \sqrt{\hbar\nu/\epsilon_0 V}$ where ν is the photon frequency. Then for both systems the atom field state evolves according to $U(t) = \exp(-iVt/\hbar)$; and one finds

$$U(t)|b,1\rangle = \cos\left(\frac{1}{2}gt\right)|b,1\rangle - i\sin\left(\frac{1}{2}gt\right)|a,0\rangle. \tag{7a}$$

Hence if $g\tau = \pi$, i.e. we have a single photon π pulse, then $|b,1\rangle \to |a,0\rangle$ for both atoms 1 and 2; thus from Eq.'s (6) and (7a) we find

$$U_1(\tau)U_2(\tau)|b_1b_2\rangle\frac{1}{\sqrt{2}}\left[|1_10_2\rangle - |0_11_2\rangle\right] = \frac{1}{\sqrt{2}}[|a_1b_2\rangle - |b_1a_2\rangle]|0_10_2\rangle. \tag{7b}$$

Likewise atoms 3 and 4 can be prepared in the singlet state $|s_{34}\rangle$ and we arrive at the double singlet state $|0,0\rangle_1 = |s_{12}\rangle|s_{34}\rangle$ without doing any post selection.

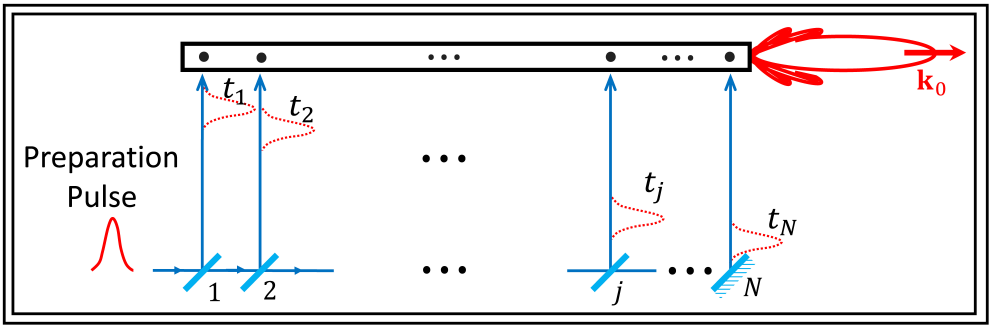

Fig. 4: Sidewise excitation of extended medium by single photon which passes through a series of beam splitters and is then directed onto an array of atoms orthogonal to the emission direction. The single photon preparation pulse is a π pulse which drives the atoms from $|b\rangle$ to $|a\rangle$.

It is also interesting to note that we can use the sidewise excitation scheme of Fig. 3 to prepare an N atom timed Dicke state without post selection. This we do by passing a single photon π pulse through a series of beam splitters (BS's) arranged as in Fig. 4. There we see a series of atoms in known fixed positions correlated with beam splitters with varying reflectance. That is, the 1st BS has a reflectance $r_1 = 1/\sqrt{N}$ and the N^{th} BS is a mirror with $r_N = 1$; each BS is selected so that a field of strength $E_{\text{in}}/\sqrt{N}$ is focused on the atoms depicted in Fig. 4. For example, for three atoms $r_1 = 1/\sqrt{3}$, $r_2 = 1/\sqrt{2}$, and $r_3 = 1$. Thus

a single photon state $|1_{\mathbf{k}_0}\rangle$ injected from the left will be split into N modes so that the N-mode photon/N atom system given by

$$|\Psi(0)\rangle = \frac{1}{\sqrt{N}} \sum_j e^{ik_0 z_j} |0_1 0_2 \cdots 1_j \cdots 0_N\rangle |b_1 b_2 \cdots b_j \cdots b_N\rangle \tag{8a}$$

evolves (for single photon π pulse excitation) according to Eq. (7) into

$$|\Psi(t)\rangle = \frac{1}{\sqrt{N}} \sum_j e^{ik_0 z_j} |b_1 \cdots a_j \cdots b_N\rangle |\{0\}\rangle \tag{8b}$$

which is the $|+\rangle_{\mathbf{k}_0}$ state of Eq. (3a). Likewise a π phase shifter placed between the $\frac{1}{2}N$ and $\frac{1}{2}N + 1$ BS will yield the $|-\rangle_{\mathbf{k}_0}$ state given by Eq. (3b).

By way of summary and open questions: the control of spontaneous emission is a problem of long-standing interest. For example, storing an excited atom in a cavity detuned from atomic resonance will slow atomic decay. Then upon switching the cavity into atomic resonance the atom will decay. This is possible on e.g. a microsecond (cavity decay time) scale. In the present scheme we can potentially hold off spontaneous emission for a much longer time; and then switch from subradiance to superradiance which can produce emission in e.g. a picosecond time scale. Sample preparation will be challenging likely involving micron cryogenic color center or nitrogen vacancy diamond quantum dots. Larger configurations involving timed Dicke states or their extensions (e.g. placing the atoms at periodic sites, such that the set $\{k_0 z_j\}$ is tailored appropriately), can also be useful and will be reported elsewhere, see also Supplement A. Other single photon subradiant states are a natural extension of the present approach. See e.g. the three sub-ensemble state depicted in Fig. B2.

$$\widetilde{|-\rangle} = \frac{1}{\sqrt{6}} \left[\sum_{j=1}^{N/3} \frac{e^{i\mathbf{k}_0 \cdot \mathbf{r}_j}}{\sqrt{N/3}} |j\rangle - 2 \sum_{j'=N/3+1}^{2N/3} \frac{e^{i\mathbf{k}_0 \cdot \mathbf{r}_{j'}}}{\sqrt{N/3}} |j'\rangle + \sum_{j''=2N/3+1}^{N} \frac{e^{i\mathbf{k}_0 \cdot \mathbf{r}_{j''}}}{\sqrt{N/3}} |j''\rangle \right]. \tag{9}$$

In particular we note that the sidewise excitation uses the single photon preparation pulses efficiently as compared to conditional excitation. It is also interesting that a semiclassical treatment fails in the scheme of Fig. 4. This will be further discussed elsewhere.

The effects of the many particle cooperative Lamb shift [18–21] have been found to be interesting in the timed Dicke single photon superradiant state. Likewise the Lamb shift in the many particle subradiant states is an interesting problem as is the Agarwal-Fano coupling between collections of single photon super- and sub-radiant states, and will be the subject of further studies. Multiphoton subradiant states (e.g. $|0,0\rangle_1$ and $|0,0\rangle_2$ of Fig. 1)

suggest interesting open questions, involving preparation of these states using classical fields. In general, single photon subradiance, its preparation and manipulation provides many open questions.

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[1] R.H. Dicke, Phys. Rev. **93**, 99 (1954).

[2] D.C. Burnham and R.Y. Chiao, Phys. Rev. **188**, 667 (1969).

[3] R. Friedberg, S.R. Hartmann, and J.T. Manassah, Phys. Rep. **7**, 101 (1973).

[4] M. Gross and S. Haroche, Phys. Rep. **93**, 301 (1982).

[5] S. Prasad and R.J. Glauber, Phys. Rev. A **61**, 063814 (2000).

[6] M.O. Scully, E.S. Fry, C.H.Raymond Ooi, and K. Wódkiewicz, Phys. Rev. Lett. **96**, 010501 (2006).

[7] I. Mazets and G. Kurizki, J. Phys. B: At. Mol. Opt. Phys. **40**, F105 (2007).

[8] E. Akkermans, A. Gero, and R. Kaiser, Phys. Rev. Lett. **101**, 103602 (2008).

[9] R. Wiegner, J. von Zanthier, and G.S. Agarwal, Phys. Rev. A **84**, 023805 (2011).

[10] W. Feng, Y. Li and S.Y. Zhu, Phys. Rev. A **89**, 013816 (2014).

[11] D.-W. Wang, H. Cai, L. Yuan, S.Y. Zhu, and R.B. Liu, Optica **2**, 712 (2015).

[12] M.O. Scully and A. Svdzinsky, Science **325**, 1510 (2009); M.O. Scully, Phys. Rev. Lett. 102, 143601 (2009).

[13] D. Pavolini, A. Crubellier, P. Pillet, L. Cabaret, and S. Liberman, Phys. Rev. Lett. **54**, 1917 (1985).

[14] R.G. DeVoe and R.G. Brewer, Phys. Rev. Lett. **76**, 2049 (1996).

[15] T. Bienaimé, N. Piovella, and R. Kaiser, Phys. Rev. Lett. **108**, 123602 (2012).

[16] B.H. McGuyer, M. McDonald, G.Z. Iwata, M.G. Tarallo, W. Skomorowski, R. Moszynski and T. Zelevinsky, Nature Physics **11**, 32 (2015).

[17] V.V. Temnov and U. Woggon, Phys. Rev. Lett. **95**, 243602 (2005).

[18] R. Friedberg and J.T. Manassah, Phys. Lett. A **372**, 2514 (2008).

[19] R. Röhlsberger, K. Schlage, B. Sahoo, S. Couet, R. Rüffer, Science **328**, 1248 (2010).

[20] J. Keaveney, A. Sargsyan, U. Krohn, I.G. Hughes, D. Sarkisyan, and C.S. Adams, Phys. Rev. Lett. **108**, 173601 (2012).

[21] Z. Meir, O. Schwartz, E. Shahmoon, D. Oron, and R. Ozeri, Phys. Rev. Lett. **113**, 193002 (2014).