Collective radiative propieties of ordered atomic chains

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We study the optical properties of an ordered atomic array when the interparticle distance is smaller than the light wavelength, for which collective effects arising from light-induced dipole-dipole interactions become relevant. In this case the scattered fields by the atoms interfere and can lead to an enhanced or suppressed spontaneous emission of photons corresponding to superradiance or subradiance, respectively. For a single excitation the system can be described in terms of collective eigenmodes, which have a modified decay rate and frequency shift compared to a single atom. By using exact diagonalization we derive the collective decay rates of an atomic array coupled to a nanophotonic waveguide and also in free space, whose atoms are transversally polarized to the atomic chain. We then study an application that takes advantage of the collective decay and subradiance arising in atomic arrays.

I. INTRODUCTION

The radiation properties of a single atom are modified in presence of other atoms due to the interaction with a common electromagnetic vacuum field. The electromagnetic vacuum can mediate dipole-dipole effective interactions between the atoms, by exchange of a virtual photon. This can lead to collective effects such as a strong enhancement or suppression of spontaneous emission of photons, leading to the well known phenomena of superradiance and subradiance [1].

Spontaneous emission of photons occurs in random directions which are often undetectable and thus, represents a loss of information hindering quantum applications based on an efficient interaction between atoms and photons. In standard treatments of atomic disordered ensembles in free space, the spontaneous emission of photons is considered to be an independent single-atom process. However, we can expect that in dense atomic media, and moreover for spatially ordered ensembles for which interference effects of the emitted fields become maximal, this assumption breaks down, and subradiance and superradiance start to be relevant.

In subradiant states, the decay is strongly suppressed and the lifetime of the atoms is increased. However, these states are by nature decoupled from optical propagating modes, what makes it difficult to access them for practical applications. Instead, selectively radiant states are those that can efficiently emit in a preferred mode that can be detected. Therefore, such states might find useful applications in the areas of quantum information processing or quantum optics.

Here we study the collective photon emission of an ordered chain of two-level atoms coupled to a nanowaveguide, and analyse a particular application where collective radiation can be exploited to create a high quality factor optical cavity with the atoms. This work is organized as follows. First we present the theoretical framework that describes the interaction between light and atoms. In particular, we present the effective Hamiltonian governing the atomic part and the relevant Green's function that describes how photons propagate in the medium. Next, we analyse the decay rate of the collective modes for three different situations. The first one, the Dicke model, a simple system of N two-level atoms with infinite range dissipative interactions. The second one, corresponds to the ordered chain coupled to a one-dimensional waveguide that mediates the interactions. The third model corresponds to an ordered chain of two-level atoms in free space. Finally, we analyse an application that exploits collective radiation of these modes. Specifically we show that two arrays of atoms coupled to a waveguide can behave as an optical cavity that reversibly exchange an excitation with an impurity atom at its center.

II. THEORETICAL FRAMEWORK

The density operator ρ is the more general object to describe an open quantum system. ρ can represent pure states and also mixed states. It is defined as:

$$\rho = \sum_{i=1}^{N} p_i \left|\Psi_i\right\rangle \left\langle\Psi_i\right| \tag{1}$$

Where Ψ is a pure state with probability p_i.

In this paper we will work with two-level atoms with ground state $|g\rangle$ and excited state $|e\rangle$. In this way and within the Born–Markov approximation [2], the evolution of ρ in time is described by *the Lindblad master equation*:

$$\partial_t \rho = -i[H,\rho]/\hbar + L[\hat{\rho}] \tag{2}$$

In absence of any external field the Hamiltonian is defined by:

$$H = \hbar\omega_0 \sum_{i=1}^{N} \hat{\sigma}_{ee}^{i} + \hbar\omega_0 \sum_{i,j=1}^{N} J^{ij} \hat{\sigma}_{eg}^{i} \hat{\sigma}_{ge}^{j} \tag{3}$$

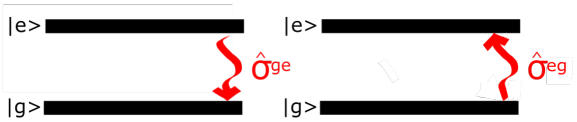

FIG. 1: Scheme of the desexcitation operator that takes the atom from the excited level to the fundamental and the excitation operator that takes the atom from the fundamental level to the excited.

Where

$$J^{ij} = -\frac{\mu_0 d_{eg}^2 \omega_{eg}^2}{\hbar} Re\left\{G\left(r, r_j, w_{eg}\right)\right\} \cdot \hat{d}_j \tag{4}$$

And the Lindbald operator:

$$L[\hat{\rho}] = \sum_{i,j}^{N} \frac{\Gamma^{ij}}{2}\left(2\sigma_j^{eg}\hat{\rho}\sigma_i^{ge} - \sigma_i^{eg}\sigma_j^{ge}\hat{\rho} - \hat{\rho}\sigma_i^{eg}\sigma_j^{ge}\right) \tag{5}$$

Where

$$\Gamma^{ij} = \mu_0 d_{eg}^2 \omega_{eg}^2 \bar{d}_j^* \cdot Im\left\{G\left(r_j, r_l, w_{eg}\right)\right\} \tag{6}$$

Where d is the electric dipole moment strength, $\hat{d}_j$ is the polarization orientation and $\hat{\sigma}^{ge}$/ $\hat{\sigma}^{eg}$ are the desexcitation/excitation operators (see Fig.1). The function $G\left(r_j, r_l, w_{eg}\right)$ is the electromagnetic Green's tensor, that depends on the particular electromagnetic environment and describes how photons propagate between two atoms.

The two last terms of the Lindblad operator Eq.(5) can be recasted in the form of an effective non-Hermitic Hamiltonian, defined as:

$$H_{\text{eff}} = \hbar\omega_0 \sum_{i=1}^{N} \hat{\sigma}_{ee}^i + \hbar\omega_0 \sum_{i,j=1}^{N}\left(J^{ij} - i\frac{\Gamma_{ij}}{2}\right)\hat{\sigma}_{eg}^i\hat{\sigma}_{ge}^j \tag{7}$$

that dictates the deterministic evolution of the atoms following the differential equation $i\partial_t\rho = (H_{\text{eff}}\rho - \rho H_{\text{eff}}^\dagger)$. Besides this evolution, the atoms can undergo quantum jumps given by the first term in the Lindblad expression. However, in our work we will be interested in the case where a single excitation is present in the system. In this case, the stochastic or quantum jumps only trivially desexcite the atoms leading them into the ground state. For most observables of interest here the physics can be properly described only with $\hat{H}_{\text{eff}}$ [3].

III. COLLECTIVE DECAY RATES IN ORDERED ATOMIC ARRAYS

A. Dicke model

The phenomena of superradiance arising in a collection of particles interacting with a common radiative bath was first introduced by Dicke in 1954 [1]. He studied the collective radiative properties of a gas of molecules when they are confined in a small volume compared to the light wavelength, and effectively can be described as being at the same real space position. This case corresponds to a constant value of the coupling constants $J^{ij} = 0$ and $\Gamma^{ij} = \Gamma_0$ in Eq.(7), for all possible pairs of interacting atoms.

As a first example, and to get some intuition, we will study how subradiant an superradiant eigenmodes arise in this simple system, for which H_{eff} can be rewritten as:

$$\hat{H}_{\text{eff}}^{\text{Dicke}} = -i\frac{\Gamma_0}{2}\sum_{i,j}^{N}\hat{\sigma}_j^{eg}\hat{\sigma}_i^{ge}, \tag{8}$$

Let us first consider only the case of two atoms. For a single excitation, the basis vectors are $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$. We can construct H_{eff}:

$$H_{\text{eff}}^{\text{Dicke}} = -i\frac{\Gamma_0}{2}\begin{pmatrix}1 & 1\\ 1 & 1\end{pmatrix} \tag{9}$$

and diagonalize it in order to find the eigenvectors and the eigenvalues:

$$\begin{aligned}&\lambda_+ = \Gamma_0 \quad \text{and} \quad \lambda_- = 0\\ &|+\rangle = \tfrac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle\right).\\ &|-\rangle = \tfrac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)\end{aligned} \tag{10}$$

This represents collective modes where the excitation is delocalized over the two atoms. The fields emitted by the two atoms perfectly constructively (destructively) interfere leading to the brightest (darkest) state with maximum (minimum) decay rate λ_+ (λ_-).

For the case of N identical atoms, we can define collective spin operators $S^\alpha = \sum_i \sigma_i^\alpha$. Taking into account that $\hat{S}^\pm = \hat{S}^x \pm i\hat{S}^y$, the Hamiltonian can be rewritten as

$$H_{\text{eff}}^{\text{Dicke}} = -i\frac{\Gamma_0}{2}\left(\hat{S}^2 - (\hat{S}^z)^2\right). \tag{11}$$

Therefore the eigenmodes of this Hamiltonian have a well defined value of the collective spin operator $\hat{S}$ and third component $\hat{S}_z$. The decay rate is then given by $\Gamma(S, S_z) = \Gamma_0 S(S+1) - S_z$. In particular, the brightest state is the state with maximum value of $S = N/2$ (maximally symmetric) and $S_z = 0$, whereas the darkest state is the total singlet state with $S = 0$ and $S_z = 0$.

B. 1D Waveguide

In this section we will work with N two-level atoms coupled to a 1D dielectric nanofiber oriented along $\hat{x}$ axis (Fig. 2). Atoms are separated by a constant distance d.

As shown in Fig. 2 the atom emits in two different ways. The decay rate Γ' refers to spontaneous emission

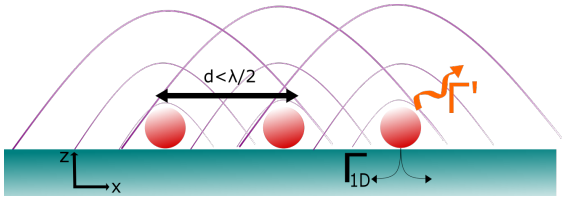

FIG. 2: Representation of the waveguide: N atoms in chain along a nanofiber in the $\hat{x}$ axis with a distance $d < \lambda/2$. Γ' and Γ_{1D} represents the singel-atom emission rates into the free space and into the waveguide, respectively.

of nonguided photons into free space modes and Γ_{1D} is the photon decay into the waveguide mode. In this case the Green's function can be approximated by:

$$G\left(r_i, r_j, w_{eg}\right) = G_0\left(r_i, r_j, w_{eg}\right) + G_{1\mathrm{D}}\left(r_i, r_j, w_{eg}\right), \quad (12)$$

Where the first term G_0 is the Green's tensor in free space, given by

$$G_0\left(r_j, r_l, w_{eg}\right) = \frac{e^{ik_0 r}}{4\pi k_0^2 r^3}\left(\left(k_0^2 r^2 + ik_0 r - 1\right)\mathbb{I}\right) + \quad (13)$$

$$\left(-k_0^2 r^2 - 3ik_0 r + 3\right)\frac{r \bigotimes r}{r^2}, \quad (14)$$

while the $G_{1\mathrm{D}}$ is the Green's function associated with the nanofiber guided mode

$$G_{1\mathrm{D}}\left(r_j, r_l, w_{eg}\right) = g\, e^{ik_{1\mathrm{D}}|z_i - z_j|}, \quad (15)$$

with g being a constant that depends on the radial distance of the atoms to the nanofiber and the exponential represents the mediated interaction of the atoms by the waveguide mode.

For the moment, we will only consider mediated interactions through the nanofiber guided mode, which means that $G = G_{1\mathrm{D}}$. In this case the effective Hamiltonian is:

$$\hat{H}_{\mathrm{eff}}^{1\mathrm{D}} = -\frac{\Gamma_{1D}}{2}\sum_{i,j} e^{ik_{1\mathrm{D}}|z_i - z_j|}\hat{\sigma}_i^{eg}\hat{\sigma}_j^{ge} \quad (16)$$

We now calculate the eigenvalues and the eigenvectors of the Hamiltonian in order to analyze the decay rate modes. Fig 3 shows the decay rate (imaginary part of the eigenvalues) with $N = 20$ for different values of d/λ.

As we can see it has a periodically behaviour. For $d/\lambda = n/2$ with $n \in \mathbb{Z}^+$ we can see a radiant state with collective decay $\Gamma = N\Gamma_0$, together with $N - 1$ completely dark states. This is due to the constructive and destructive interference between the electromagnetic waves emitted by the atoms, respectively.

If all the atoms are in phase, for n even the exponential turns $+1$ and we have a constructive interference. For n odd the exponential turns $(-1)^{|i-j|}$ and we have a destructive interference (see Fig.4). Therefore a maximally bright state corresponds to a superposition of excitations between the different atoms that are in phase (out of phase, with relative phase π) for n even (odd).

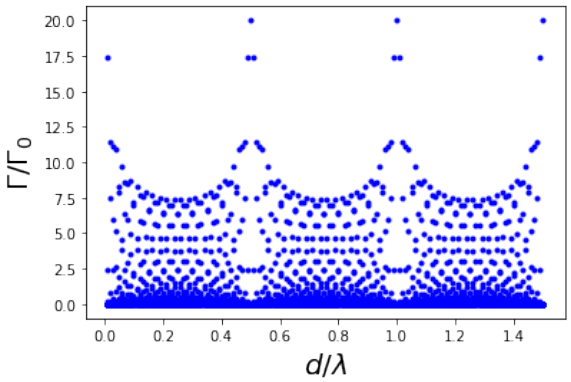

FIG. 3: Γ/Γ_0 (decay rate) of the N modes for different values of d/λ for a chain of $N = 20$ atoms coupled to a waveguide.

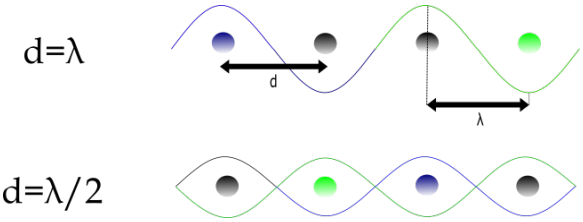

FIG. 4: Representacion of the electromagnetic waves of the atoms with n even ($d/\lambda_0 = 1$, construcitve) and with n odd ($d/\lambda_0 = 1/2$, destructive),

C. Free space: subradiant states

In this section we will apply the spin model to a one-dimensional ordered chain of N two-level atoms in free space. The polarization of the atoms is considered to be transversal with respect to the axis defined by the chain. In this conditions we can treat the position of atoms as classical points $r_{ij} = r_i - r_j = r$. By replacing this in the expression of the Green's function (14) and inserting it in the effective Hamiltonian (7) we arrive at:

$$\hat{H}_{\mathrm{eff}}^0 = -\frac{3\pi\Gamma_0}{k_0}\sum_{i,j}^{N}\frac{e^{ik_0 r}}{4\pi k_0^2 r^3}\left(\left(k_0^2 r^2 + ik_0 r - 1\right)\mathbb{I} + \left(-k_0^2 r^2 - 3ik_0 r + 3\right)\frac{r \bigotimes r}{r^2}\right)\hat{\sigma}_i^{eg}\hat{\sigma}_j^{ge} \quad (17)$$

where $\Gamma_0 = |d|^2 k_0^3/3\pi\hbar\epsilon_0$ is the rate of spontaneous emitted photons, d the lattice constant, $k_0 = \omega/c$ is the wave number of the atomic transition and $r \bigotimes r$ is a tensor product that depends on the direction of the polarization and the particles position. More specifically, in our system (1D chain), the atoms are in the z axis so $r = z_i - z_j$ and x is the direction of the polarization. We have to take into count that we have a term that goes like $1/(z_i - z_j)^\alpha$, so for $r_i = r_j$ we have a discontinuity. We can fix it by doing an expansion of the Green's function in the limit $r_i \to r_j$ so that $\rho^2 G_0(r_i \to r_j) = -i\Gamma_0/2$ [5].
We can now apply all this formalism to a real system.

We fix the number of atoms to $N = 50$ and analyze how the decay varies with d/λ_0. For each d/λ_0 we diagonalize H_{eff} and compute the eigenvalues. The decay rate (the imaginary part of the eigenvalues) has been calculated as a function of Γ_0 so that the result is expressed in terms of the single-atom spontaneous emission rate. The results are shown in Fig. 5.

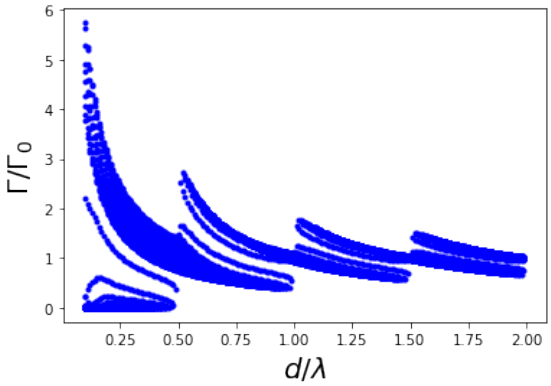

FIG. 5: Γ/Γ_0 (decay rate) of the N modes for different values of d/λ_0 for a chain of N = 50 atoms in 1D free space with transverse polarization.

We can see that for large d/λ_0 the collective decay rate $\Gamma \to \Gamma_0$ because this limit corresponds to a system with effectively independent atoms and therefore the decay rate is the spontaneous emission rate of a single atom Γ_0. In the other hand, for decreasing d/λ_0 we observe a periodical behaviour due to the constructive or destructive interference of the fields emitted by the atoms. Finally we notice an interesting behaviour for $d/\lambda_0 < 1/2$. In this region there exist collective modes with $\Gamma/\Gamma_0 \ll 1$, which are strongly subradiant.

IV. AN APPLICATION: CAVITY QED WITH ATOMIC MIRRORS

In this section we will show that it is possible to construct an effective high finesse cavity coupled to an impurity atom by taking advantage of the collective radiative properties of ordered atomic arrays in free space studied in the previous section. Let us consider a chain of atoms coupled to the waveguide that can spontaneously emit a photon into the waveguide mode at rate Γ_{1D}. If the atoms are separated by $d = \lambda/2$, and as shown before, the system possesses a single bright mode and can act as an atomic mirror, perfectly reflecting light. By coupling two of these atomic mirrors to an additional impurity atom placed at a particular distance (cavity QED configuration as shown in Fig. 6), we will observe vacuum Rabi oscillations where an excitation is reversibly exchanged between the impurity atom and the rest of the chain (see Fig. 7).

If the two mirrors are separated by a distance which is

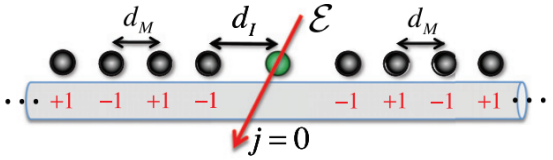

FIG. 6: Representation of the cavity QED configuration extracted from [4]. As we can see there are two chains of atoms, one on each side separated by a distance $d = \lambda/2$, therefore they act as mirrors. Between these two chains we have the impurity atom. Finally ε is an additional term for guiding the impurity ato

$(2n+1)\lambda_{1D}/2$ ($n \in \mathcal{Z}^+$), and the impurity is exactly at the center between the two mirrors, only a single collective mode of the atomic mirrors (called the cavity mode) with associated creation operator

$$S^\dagger_{\text{cav}} = \frac{1}{\sqrt{N_A}} \sum_{j>0} (-1)^j \left(\hat{\sigma}^{eg}_j + \hat{\sigma}^{eg}_{-j}\right) \tag{18}$$

couples to the impurity. Then, the effective Hamiltonian reduces to:

$$\hat{H}^{\text{QED}}_{\text{eff}} = g\left(\hat{\sigma}^{ge}_I S^\dagger_{\text{cav}} + \hat{\sigma}^{eg}_I S_{\text{cav}}\right), \tag{19}$$

with $g = \Gamma_{1D}\sqrt{N_A}/2$ and $\hat{\sigma}^{eg}_I$ ($\hat{\sigma}^{ge}_I$) the creation (annihilation) operator of an excitation of the impurity.

With this atomic configuration, we study first the dynamic evolution of the excited state population of the impurity by starting with the initial state where all atoms are in the ground state, except of the impurity. In absence of any additional decay rather than the one into the fiber mode (that is, the system evolving under under $\hat{H}^{1D}_{\text{eff}}$), we observe the so-called Rabi oscillations with damping given by Γ_{1D} (the decay into the fiber). This is shown in Fig.7(a) (blue solid line). This result is in stark contrast to the case where atoms are spontaneously emitting photons into free space in an independent fashion, defined the following effective Hamiltonian:

$$\hat{H}^{\text{indep}}_{\text{eff}} = -i\Gamma_0/2 \sum_{j=1}^{N} \hat{\sigma}^j_{ee}. \tag{20}$$

In this case, the excited state population of the impurity simply decays exponentially with time, as shown by the red dashed curve in Fig.7(a).

If we now consider independent atom decay into free space modes on top of the nanofiber guided interaction, that is, we let the system evolve under $\hat{H}_{\text{eff}} = \hat{H}^{1D}_{\text{eff}} + \hat{H}^{\text{indep}}_{\text{eff}}$ we observe that the Rabi oscillations are destroyed. This is shown in Fig.7(b) (red dashed curve). We finally study the case where spontaneous emission into free space is also collective, corresponding to the case where $d < \lambda_0/2$, where λ_0 is the wavelength associated with the atomic transition. In this case, for which the system evolves under $\hat{H}_{\text{eff}} = \hat{H}^{1D}_{\text{eff}} + \hat{H}^{0}_{\text{eff}}$ we observe that Rabi oscillations can be recovered as the atoms are

in a subradiant state with respect to free space emission.

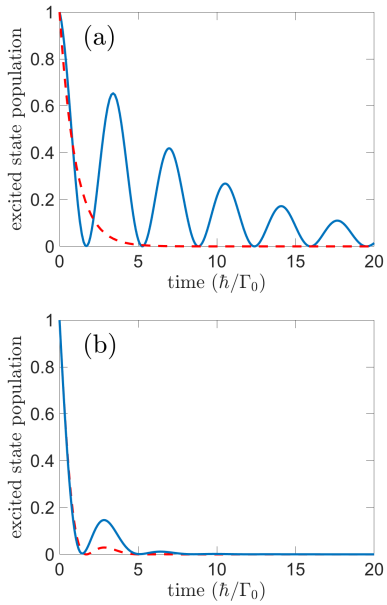

FIG. 7: Excited state population of the impurity atom as a function of time for different cases: (a) Decay only into the waveguide mode (solid blue) and only independent decay into free space modes (dashed red). In the former there is a reversible exchange of the excitation between the mirrors and the impurity. (b) Collective decay into the fiber and independent decay into free space modes (dashed red), and collective decay into the fiber and into free space (blue solid). In presence of collective decay the Rabi oscillations are recovered. ($\Gamma_{1D} = \Gamma_0/4$, $\lambda_{1D} = 2d$, $\lambda_0 = 2d$, $N = 50$).

V. CONCLUSION

We have shown how the radiative properties, such as the decay due to spontaneous emission of photons, of a collection of atoms are modified when they are coupled to a common electromagnetic bath. We started finding the equations that define the interactions of light and matter and its evolution. We found the Lindblad master equation with the corresponding Hamiltonian and Lindbald operator. We rewrote the Hamiltonian as a function of the coherent and dissipative interactions. Finally we defined the effective Hamiltonian that dictates the deterministic evolution of the atoms. We built the whole theoretical model that has allowed us to know the collective behaviors of atoms.

Deriving the Hamiltonian from the different situations posed and using computational methods we have been able to know which modes of interaction the atoms develop. Depending on how the atoms are arranged and according to the parameters of the wavelength we have observed superadiance (bright state) or subradiance (dark state). For the waveguide case we observed the emergence of perfectly dark states and a single bright mode for $d/\lambda_0 = n/2$, with n being an integer. On the other hand, for the case of free space we also observe subradiance ($\Gamma \ll \Gamma_0$) when the interparticle separation is smaller than half of the light wavelength.

Using the collective radiative properties that arise in these systems we propose a protocol with two ordered chains of atoms and an impurity atom at its center acting as if they were two mirrors of a cavity coupled to a single atom. With the appropriate spatial arrangement it is possible to observe a reversible exchange of an excitation between the mirror atoms and the impurity. If the atoms can only decay into the waveguide mode, the oscillations are maximal and damped with the rate Γ_{1D}. For atoms that are separated by large distances compared to the atomic transition wavelength (i.e., $d/\lambda_0 > 1/2$) spontaneous emission into free space can destroy these oscillations. However, we have shown that if the particles are placed at closer distances (i.e., $d/\lambda_0 < 1/2$), for which subradiant modes start to play a role, the oscillations are recovered.

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[1] H Dicke, Coherence in Spontaneous Radiation Processes, Phys. Rev., Vol.93, p.99 1954
[2] C. W. Gardiner et al. "Quantum noise: a handbook of Markovian and non-Markovian quantum stochastic methods with applications to quantum optics", Springer Science & Business Media 2004.
[3] OM T Manzoni et al. *Optimization of photon storage fidelity in ordered atomic arrays* 2018 New J. Phys. 20 083048
[4] D E Chang et al. *Cavity QED with atomic mirrors* 2012 New J. Phys. 14 063003.
[5] A. Asenjo-Garcia, et al. *Exponential Improvement in Photon Storage Fidelities Using Subradiance and "Selective Radiance" in Atomic Arrays*,PHYSICAL REVIEW X 7, 031024 (2017)
[6] M T Manzoni et al. *Optimization of photon storage fidelity in ordered atomic arrays* 2018 New J. Phys. 20 083048