Non-Markovian Collective Emission from Macroscopically Separated Emitters

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We study the collective radiative decay of a system of two two-level emitters coupled to a one-dimensional waveguide in a regime where their separation is comparable to the coherence length of a spontaneously emitted photon. The electromagnetic field propagating in the cavity-like geometry formed by the emitters exerts a retarded backaction on the system leading to strongly non-Markovian dynamics. The collective spontaneous emission rate of the emitters exhibits an enhancement or inhibition beyond the usual Dicke superradiance and subradiance due to self-consistent coherent time-delayed feedback.

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Introduction.—Long-distance interactions are a central tenet of many quantum systems and processes, including large-scale quantum networks, distributed quantum sensing, and information processing [1–5]. When the separations between emitters become comparable to the coherence length of the photons mediating their interaction, interference properties of the electromagnetic (EM) field can be modified due to retardation. In such cases, the backaction of the EM field on the emitters leads to coherent time-delayed feedback on the system dynamics [6,7], thus rendering it non-Markovian [8–11].

Non-Markovian open system dynamics can have a variety of physical origins such as structured bath spectral densities, strong system-bath couplings, low temperatures, or initial system-bath correlations among others [9,10,12–15]. The effects of non-Markovianity have been investigated in collective atomic states in the context of structured reservoirs [16–20] and in the strong-coupling regime [21]. Furthermore, delay-induced non-Markovian dynamics has been previously shown in the context of spontaneous emission of single atoms [22–30], bound states in continuum (BIC) of the EM field [31–35], and entanglement generation in emitters coupled to waveguides [4,36].

Additional insights into non-Markovian effects in such a regime can be gained from studying the simple, yet rich quantum optics phenomenon of collective spontaneous emission of two two-level emitters. Cooperative effects in spontaneous emission have an extensive historical background [37–40], and have been experimentally observed across a range of physical systems [41–49]. While the influence of retardation on these effects has been previously studied in Refs. [50,51], the non-Markovian dynamics emerging in macroscopically delocalized collective systems is yet unexplored.

In this Letter we study the collective radiative dynamics of a pair of macroscopically separated emitters and show that it exhibits non-Markovian features caused by self-consistent coherent time-delayed feedback. We specifically consider here the emitters are prepared in a superradiant or subradiant electronic state, and present an exact analytical solution of the dynamics of the collective spontaneous emission. We demonstrate that the retarded backaction of the EM field on the emitters can lead to a further enhanced (inhibited) spontaneous emission rate for superradiant (subradiant) states beyond the usual Dicke superradiance (subradiance) [37,38].

We consider two two-level emitters coupled to a waveguide are separated by a distance d comparable to the coherence length $\sim v_g/\gamma$ of a spontaneously emitted photon, with v_g being the group velocity of the field and γ the spontaneous emission rate of the individual emitters (see Fig. 1). To gain an intuitive understanding of the non-Markovian nature of this system, consider the following apparent "superradiance paradox": Assume that the distance d between two emitters prepared in a superradiant state is smaller than the coherence length of an independently emitted photon, but larger than that of a superradiant photon, $v_g/\gamma > d > v_g/(2\gamma)$. Given that superradiance is an interference effect, one would expect to observe superradiant emission if there is no way to distinguish which atom emitted the field [52]. Now if the emitters radiate collectively, with an emission rate 2γ, then the coherence length of the emitted photons $[v_g/(2\gamma)]$ is too short to allow for the fields radiated by the two emitters to interfere, suggesting that they should have emitted independently. On the other hand, if we assume that they do emit independently, then the coherence length of the emitted photons (v_g/γ) is long enough that there should be interference and as a result the emitters should emit at the superradiant rate of 2γ instead. This seeming paradox points to the failure of the Markov approximation: the conventional notion of an exponential decay defining the photon coherence length is no longer valid, and it is necessary instead to consider a full non-Markovian treatment of the system dynamics.

Formal development.—The total Hamiltonian for the emitters + field system is $H = H_E + H_F + H_{\rm int}$, where $H_E = \hbar\omega_0 \sum_{m=1,2} \hat{\sigma}_+^{(m)} \hat{\sigma}_-^{(m)}$ is the free Hamiltonian for the emitters of resonance frequency ω_0, with $\hat{\sigma}_+^{(m)}$ is the creation operator of an excitation in the mth emitter; $H_F = \hbar\omega \int_0^\infty \mathrm{d}\omega[\hat{a}^\dagger(\omega)\hat{a}(\omega) + \hat{b}^\dagger(\omega)\hat{b}(\omega)]$ is the free Hamiltonian for the field, with $\hat{a}(\omega)$ and $\hat{b}(\omega)$ the annihilation operators for the right- and left-propagating field modes of the waveguide, respectively; and $H_{\rm int}$ is the emitter-field interaction Hamiltonian.

We proceed by making the electric-dipole and rotating-wave approximations (RWA) and expressing the emitters-field interaction Hamiltonian in the interaction picture with respect to the total free Hamiltonian $H_E + H_F$ as

$$H_{\rm int} = \hbar \sum_{m=1}^{2} \int_0^\infty \mathrm{d}\omega [g(\omega)\hat{\sigma}_+^{(m)} \{\hat{a}(\omega) e^{i\omega x_m/v_g} + \hat{b}(\omega) e^{-i\omega x_m/v_g}\} e^{-i(\omega-\omega_0)t} + \mathrm{H.c.}], \tag{1}$$

where $g(\omega)$ is the atom-field coupling strength [53,56]. To isolate the non-Markovian behavior arising from the retardation effects from that due to a structured reservoir,

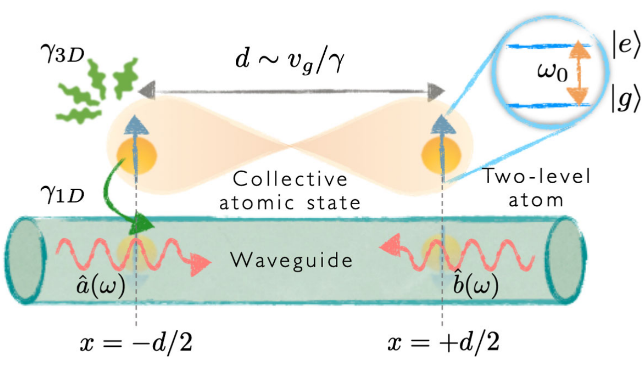

FIG. 1. Two two-level emitters prepared in a collective state coupled to an optical waveguide. The emitters are located at positions $x_{1,2} = \pm d/2$, with d comparable to the coherence length $\sim v_g/\gamma$. The rates γ_{3D} and γ_{1D} refer to the emitter spontaneous emission rates into free space and guided modes, respectively. The mode operators $\hat{a}(\omega)$ and $\hat{b}(\omega)$ refer to annihilation operators for the right- and left-propagating waveguide modes, respectively.

we assume a flat spectral density of the field modes around the resonance of the emitters such that $g(\omega) \approx g(\omega_0)$.

Assuming that the total emitters plus field system is initially prepared in the single-excitation manifold, and considering that in the RWA the Hamiltonian preserves the total number of excitations, the state at time $t > 0$ is

$$|\Psi(t)\rangle = \sum_{m=1}^{2} c_m(t)\hat{\sigma}_+^{(m)}|g,g,\{0\}\rangle + \int_0^\infty \mathrm{d}\omega[c_a(\omega,t)\hat{a}^\dagger(\omega) + c_b(\omega,t)\hat{b}^\dagger(\omega)]|g,g,\{0\}\rangle, \tag{2}$$

where c_m and $c_{a,b}(\omega)$ are the excitation amplitudes for the mth emitter and the guided field modes with frequency ω, respectively, and $|g,g,\{0\}\rangle$ is the ground state of the total system, with $|\{0\}\rangle$ the field vacuum state. Tracing out the field modes, evolution of emitter excitation amplitudes is given by [61]

$$\dot{c}_m(t) = -\frac{\gamma}{2}[c_m(t) + \beta c_n(t - d/v_g)\Theta(t - d/v_g)e^{i\phi_p}] \tag{3}$$

for $m \neq n$, where $\phi_p \equiv k_0 d = 2p\pi$ is the field phase difference upon propagation, which we assume to be an integer multiple of 2π, $\gamma \equiv \gamma_{1D} + \gamma_{3D}$ is the total spontaneous emission rate, and $\gamma_{1D} = \beta\gamma \equiv 2\pi|g(\omega_0)|^2$ is the spontaneous emission rate into the waveguide, with β the coupling efficiency of the emitters to the waveguide. The second term in Eq. (3) represents the retarded backaction of the other emitter via the field with a delay $t = d/v_g$.

For emitters initially in the superradiant or subradiant states $|\Psi_{\rm sub}^{\rm sup}\rangle \equiv (1/\sqrt{2})(|eg\rangle \pm |ge\rangle)$ [62], one can write the Laplace transformed coefficients $\tilde{c}_m(s) \equiv \int_0^\infty \mathrm{d}t\, e^{-st} c_m(t)$ as

$$\tilde{c}_{\rm sup}(s) = \frac{1}{\sqrt{2}\gamma[\tilde{s} + 1/2 + \beta e^{-\eta\tilde{s}}/2]}, \tag{4}$$

$$\tilde{c}_{\rm sub}(s) = \frac{1}{\sqrt{2}\gamma[\tilde{s} + 1/2 - \beta e^{-\eta\tilde{s}}/2]}, \tag{5}$$

where $\tilde{s} \equiv s/\gamma$ and $\eta \equiv d\gamma/v_g$ is the separation between the emitters normalized by the photon coherence length. Here $\tilde{c}_{\rm sup} = \tilde{c}_1^{\rm sup} = \tilde{c}_2^{\rm sup}$ and $\tilde{c}_{\rm sub} = \tilde{c}_1^{\rm sub} = -\tilde{c}_2^{\rm sub}$ are the Laplace space probability amplitudes for the superradiant and subradiant cases, respectively [63].

Consider next the case where the emitters are slightly separated, $\eta \ll 1$. Up to linear terms in η

$$\tilde{c}_{\rm sub}^{\rm sup}(\tilde{s}) \approx \frac{1}{\sqrt{2}[s(1 \mp \beta\eta/2) + \gamma/2(1 \pm \beta)]}, \tag{6}$$

which yields an effective spontaneous emission rate

$$\gamma_{\rm sub}^{\rm sup} \approx \frac{1 \pm \beta}{1 \mp \beta\eta/2}\gamma. \tag{7}$$

For a small but finite delay $0 < \eta \ll 1$, this can potentially exceed the usual Dicke superradiant emission rate of 2γ for $\beta = 1$. Also, for a subradiant state with an imperfect coupling ($\beta < 1$), the effective decay for slightly separated emitters can be slower than that for coincident ones. This surprising enhancement and inhibition of the collective spontaneous emission can be attributed to a stimulated emission as the correlated field emitted from one of the emitters interferes with that from the other [64]. The separation dependence of the collective emission rate, in addition to the phase difference, demonstrates the influence of retardation on the interference.

We now consider the general case of arbitrarily separated emitters, for which we present an exact analytical solution of the equations of motion (3) based on a well-developed mathematical treatment of delay differential equations (see [65] and the Supplemental Material (SM) [66] for details). The general expression for the excitation amplitudes of the emitters is

$$c_{\rm sub}^{\rm sup}(t) = \frac{1}{\sqrt{2}}\sum_{n\in\mathbb{Z}}\alpha_n^{(\pm)}e^{-\gamma_n^{(\pm)}t/2}, \tag{8}$$

where $\alpha_n^{(\pm)} \equiv \{1 + W_n[\mp (\eta/2)\beta e^{\eta/2}]\}^{-1}$ and the effective decay rate $\gamma_n^{(\pm)} \equiv \gamma\{1 - 2W_n[\mp (\eta/2)\beta e^{\eta/2}]/\eta\}$, with $W_n(x)$ the nth branch of the Lambert W function, which is commonly used to describe systems that exhibit time-delayed feedback [61,65]. We now discuss the consequences of this analytical solution, which is the main result of this work.

Results.—Consider first the dynamics of a superradiant initial state. From Eq. (8) and the properties of the Lambert-W function, one finds that the superradiant solution has imaginary exponents for $\eta > \eta_c$, where we have introduced the normalized critical distance $\eta_c \equiv 2W_0[1/(e\beta)]$ [61]. Thus for $\eta \geq \eta_c$, the atomic excitation amplitudes exhibit oscillations as the atoms decay to their ground state. These can be understood in terms of a field wavepacket bouncing back and forth between the emitters [50,66]. For $\beta = 1$ this occurs for separations $d > 0.56 v_g/\gamma$, as shown in Fig. 2.

For separations $\eta < \eta_c$ the emitters radiate independently until a time $\gamma t = \eta$ and collectively afterwards, with an instantaneous decay rate given by

$$\gamma_{\rm inst} = \gamma\left[1 - \frac{W_0[-(\eta/2)\beta e^{\eta/2}]}{\eta/2}\right]. \tag{9}$$

For a given value of β, this rate reaches a maximum $\gamma_{\rm inst}^{\rm max}$ when the normalized emitter separation equals its critical value $\eta = \eta_c$, with $\gamma_{\rm inst}^{\rm max}/\gamma = 1 - \{[W_0(-1/e)]/W_0[1/(e\beta)]\}$, as shown in Fig. 3. In the absence of losses and for perfect emitter-waveguide coupling efficiency ($\beta = 1$), the maximum instantaneous spontaneous emission rate is $\gamma_{\rm inst}^{\rm max}/\gamma \approx 4.59$, in stark contrast with superradiant emission in Markovian systems.

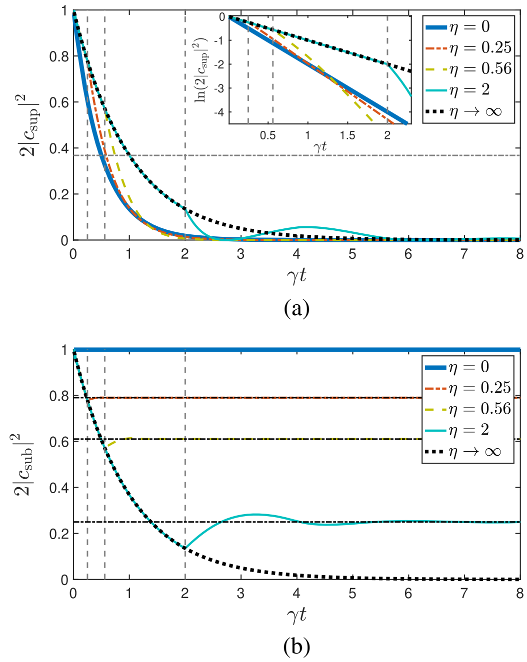

FIG. 2. Emitters excitation probabilities as a function of time and for different separations η for initial (a) superradiant and (b) subradiant states, assuming perfect coupling efficiency ($\beta = 1$). Solid and dotted curves depict the dynamics for $\eta = 0$ and $\eta = \infty$, respectively. For intermediate emitter separations, the emitters decay independently with a rate γ until $\gamma t = \eta$ (indicated by the dashed vertical lines), and collectively afterwards. For the critical separation $\eta \approx \eta_c \approx 0.56$, we observe an instantaneous superradiant spontaneous emission rate of $\gamma_{\rm inst} \approx 4.59\gamma$. The $1/e$ value of the initial emitters excitation probability is reached first for coincident emitters (depicted by the gray horizontal dashed-dotted line). In the subradiant case the emitter excitation probability reaches the asymptotic value $(1 + \eta/2)^{-2}$, shown by the horizontal dashed-dotted lines.

In the case of a subradiant initial state, for $\beta = 1$, the steady state of the dynamics corresponds to a BIC [33]. The probability of reaching the BIC, starting initially in the subradiant state of the atoms, is given by $|\langle\Psi(t\to\infty)|\Psi_{\rm BIC}\rangle|^2 = 1/(1 + \eta/2)$ [73]. The total probability of the emitters being excited in the steady state is $|c_{1,2}^{\rm sub}(\infty)|^2 \to \{1/[2(1 + \eta/2)^2]\}$; see Fig. 2(b). We also note that for an initial subradiant state with a delay of $\eta \approx 0.8$, it is possible to achieve a maximal emitter-field steady state entanglement [66].

It is also instructive to explore the cooperative nature of the atom-field dynamics from the perspective of the emitted field intensity $I(x,t) \propto \langle\Psi(t)|\hat{E}^\dagger(x,t)\hat{E}(x,t)|\Psi(t)\rangle$, where the electric field operator is $\hat{E}(x,t) \propto$

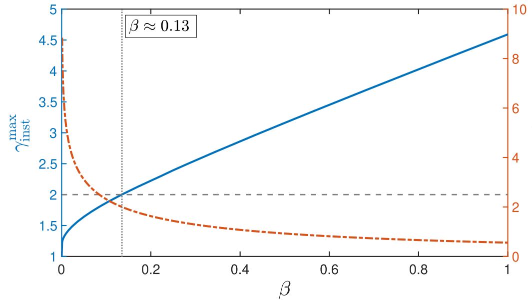

FIG. 3. Instantaneous decay rate $\gamma_{\rm inst}^{\rm max}$ (blue solid line) and associated critical emitter separation η_c (red dashed-dotted line) as a function of the emitter-waveguide coupling efficiency β for an initial superradiant state. The horizontal dashed line depicts an instantaneous collective emission rate of 2γ, which corresponds to a coupling efficiency of $\beta \approx 0.13$. This illustrates that the collective emission rate of 2γ of usual Dicke superradiance can be exceeded for sufficiently large emitter-waveguide coupling efficiency and appropriate emitter separations.

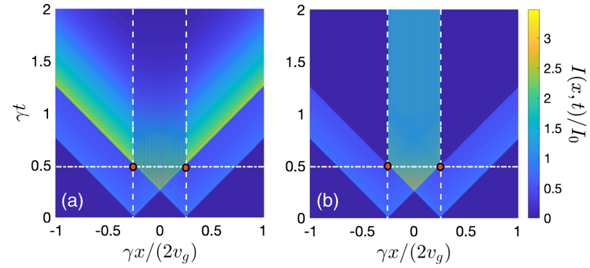

FIG. 4. Normalized field intensity as a function of position and time for $\eta = 0.5$ and (a) superradiant and (b) subradiant initial state. The positions of emitters at $\gamma x_{1,2}/(2v_g) \approx \pm 0.25$ are depicted by the vertical dashed line. As the field from one of the emitters reaches the other at $\gamma t = \eta$ (dashed-dotted line), their interference results in either a collective radiation burst or reflection of the field into the cavity formed by the emitters.

$\int_0^\infty dk[\hat{a}(k)e^{ikx} + \hat{b}(k)e^{-ikx}]e^{i\omega t}$ and $|\Psi(t)\rangle$ the state of the system (see [66] for details). Figure 4 shows that the fields emitted by the two emitters in the superradiant (subradiant) case interfere constructively (destructively) when the light cones of the two emitters reach each other. Thereafter, depending on their relative phase they produce an interference pattern that is either constructive, leading to a collective "superduperradiant" burst with an instantaneous emission rate greater than 2γ, or destructive, resulting in the a perfect reflection of the field into the optical cavity created by the two atoms. The nonexponential decay of the emitters is an unambiguous signature of the non-Markovian evolution of the system, a result of the self-consistent backaction of the EM field bath, which is accounted for by a departure from the usual Lindblad dynamics. We further quantify the non-Markovianity of the system in the Supplemental Material [68], which shows that the system is non-Markovian for any value of η, approaching a Markovian behavior for $\eta \to 0$.

Noting that in the presence of delay the instantaneous collective decay rate can exceed that of standard Dicke superradiance, one might wonder if the total collective emission into the waveguide also gets enhanced. An important figure of merit to quantify the collective nature of the system in this regard is its cooperativity $\mathcal{C} \equiv \gamma_{\rm in}/\gamma_{3D}$ [74], such that $\gamma_{\rm in} = \lim_{t\to\infty}\int_0^\infty d\omega[|c_a(\omega,t)|^2 + |c_b(\omega,t)|^2]$ is the fraction of the field emitted into the waveguide and $\gamma_{3D} = \gamma(1-\beta)$ is the fraction of the field that escapes out to the nonguided modes [75]. This can be evaluated as [66]

$$\mathcal{C}_{\rm sub}^{\rm sup} = \frac{\beta}{1-\beta}\sum_{m,n}\frac{\alpha_n^{(\pm)}\alpha_m^{(\pm)*}}{\gamma_n^{(\pm)} + \gamma_m^{(\pm)*}} [2 \pm \{e^{-\eta\gamma_n^{(\pm)}/(2\gamma)} + e^{-\eta\gamma_m^{(\pm)*}/(2\gamma)}\}.] \qquad (10)$$

For $\eta > 0$, the cooperativity for a superradiant state is reduced compared to that of coincident emitters ($\eta = 0$) as the total collective emission into the guided modes decreases with the emitter separation. In contrast, for an antisymmetric state we find an enhanced emission into the waveguide as η is increased. This is due to the emission of the field into guided modes by the individual emitters until $\gamma t = \eta$, before they start acting collectively [see Fig. 4(b)]. Given that cooperativity is an important figure of merit in quantum information applications, this result illustrates that retardation effects need to be carefully considered in quantum network protocols based on long distance emitters [76].

Summary and outlook.—We have shown that the collective radiative decay of two emitters coupled to a one-dimensional waveguide is subject to non-Markovian modifications due to the time-delayed backaction of the electromagnetic field upon the emitters. When prepared in a superradiant initial state they can exhibit time-dependent decay rates that can instantaneously surpass the standard Dicke superradiance rate. The system also allows for long-lived subradiant states characterized by a bound state in the field trapped in the region between the emitters. These effects can be understood as a combination of Dicke superadiance or subradiance and a retardation of the field wavepacket where the electromagnetic field senses its boundary conditions with a significant delay.

A key parameter for characterizing the dynamics is the emitter separation relative to the photon coherence length $\eta \equiv d\gamma/v_g$. It captures the combined physical origin of non-Markovian behavior, as an appreciable value of η can be achieved by increasing the emitter separation d, but also by increasing the system-environment coupling as in [36] or by exploiting slow group velocities achievable in the presence of a band gap or near a band edge [20]. Importantly, as η is increased the system dynamics requires keeping track of field correlation functions of increasing order. We note that the non-Markovianity in this case arises explicitly due to retarded backaction effects, despite having a flat spectral density for the bath.

Experimental observations of these effects could be realized across a number of platforms, including quantum dots in photonic waveguides [48], atoms near optical nanofibers [47,77,78], and superconducting qubits coupled by coplanar waveguides [79,80]. Table I in the SM [66] summarizes experimental parameters accessible so far. For a system of atoms coupled to nanofibers, values of $\eta \sim 1$ have already been realized [78]. Given the rapid experimental progress in all these platforms, the retarded collective effects studied here can become relevant in the near future.

With the emerging possibility of preparing collective dipoles subject to internal retardation effects and observing their associated complex dynamics in sharp contrast with the more familiar case of emitters confined in subwavelength regions, our work adds a new intricacy that has been little explored in the past. Given that the enhancement in the retarded collective decay of two emitters relies on pairwise time-delayed feedback, it will be interesting to determine the scaling of these effects with the number of emitters. We also note that similar dynamics can arise in a system of linear oscillators [81], indicating that such collective retarded dissipation should be observable in classical systems as well. It then would be interesting to extend the present dynamics from the single-excitation case considered here to multiple excitations, where one can observe genuinely quantum non-Markovian effects, such as the phenomenon of superfluorescence [82] with retardation, where all the emitters decay collectively from a fully excited state.

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