Many-Body Signatures of Collective Decay in Atomic Chains

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Fully inverted atoms placed at exactly the same location synchronize as they deexcite, and light is emitted in a burst (known as "Dicke's superradiance"). We investigate the role of finite interatomic separation on correlated decay in mesoscopic chains and provide an understanding in terms of collective jump operators. We show that the superradiant burst survives at small distances, despite Hamiltonian dipole-dipole interactions. However, for larger separations, competition between different jump operators leads to dephasing, suppressing superradiance. Collective effects are still significant for arrays with lattice constants of the order of a wavelength, and lead to a photon emission rate that decays nonexponentially in time. We calculate the two-photon correlation function and demonstrate that emission is correlated and directional, as well as sensitive to small changes in the interatomic distance. These features can be measured in current experimental setups, and are robust to realistic imperfections.

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Collective effects in the interaction between light and matter have attracted interest since the seminal work of Dicke in the 1950s, who studied the problem of photon emission by many atoms at identical locations [1]. In this purely dissipative scenario, the atomic dipoles become phase locked during their decay and emit collectively. This leads to an initial increase in the photon emission rate—the famous "superradiant burst" or "superfluorescence"—rather than the typical exponential decay for independent atoms.

Dicke's scenario ignores coherent dipole-dipole interactions between atoms, which are relevant for finite interatomic distances and have been predicted to wash out superradiant decay [2,3]. However, signatures of collective behavior persist even in systems of size much larger than the resonance wavelength. For example, theoretical studies of ordered arrays of emitters have shown the existence of extremely subradiant (i.e., dark) few-excitation states [4–12], as well as directional collective emission [4,13–21].

Recent experimental realizations of ordered atomic arrays, both in optical lattices [22–26] and tweezer arrays [27–33], open the door for investigation of these predictions. These platforms have already allowed for the demonstration of a two-dimensional atomic mirror [26] and the measurement of collective frequency shifts in a one-dimensional (1D) atomic array [33]. Current experimental capabilities offer the possibility of measuring statistics of the emitted photons. This raises the question of whether collective decay imprints correlations on the emitted photons. This would allow for the potentially tunable generation of nonclassical states of light (and maybe of a superradiant laser [34,35]), critical for quantum technologies. Conversely, connecting the correlations in the light back to the atomic quantum state would offer a unique light-based probe to characterize these dissipative many-body systems.

Here, we make an important step in this direction by investigating collective decay and superradiance in mesoscopic 1D ordered arrays. We find that a superradiant burst survives for short interatomic distances ($d \lesssim \lambda_0/4$). We use a quantum jump approach to connect the sequence of collective jumps to the statistics of the light emitted. We show that strong spatial correlations between emitted photons are imparted by the collective decay and that they persist even for $d \sim \lambda_0$.

We consider N atoms of resonance frequency ω_0, arranged in an ordered chain along the z-axis with lattice constant d, as shown in Fig. 1(a). Interactions between atoms are obtained by tracing out the electromagnetic field under a Born-Markov approximation [36,37]. The atomic density matrix, ρ, evolves under the master equation

$$\dot{\rho} = -\frac{i}{\hbar}[\mathcal{H}, \rho] + \sum_{i,j=1}^{N} \frac{\Gamma^{ij}}{2}(2\hat{\sigma}^j_{ge}\rho\hat{\sigma}^i_{eg} - \rho\hat{\sigma}^i_{eg}\hat{\sigma}^j_{ge} - \hat{\sigma}^i_{eg}\hat{\sigma}^j_{ge}\rho), \tag{1}$$

where the Hamiltonian is

$$\mathcal{H} = \hbar\sum_{i=1}^{N}\omega_0\hat{\sigma}^i_{ee} + \hbar\sum_{i,j=1}^{N} J^{ij}\hat{\sigma}^i_{eg}\hat{\sigma}^j_{ge}. \tag{2}$$

Here, $\hat{\sigma}^i_{ge} = |g_i\rangle\langle e_i|$ is the atomic coherence operator, with $|e_i\rangle$ and $|g_i\rangle$ the excited and ground states of the cycling

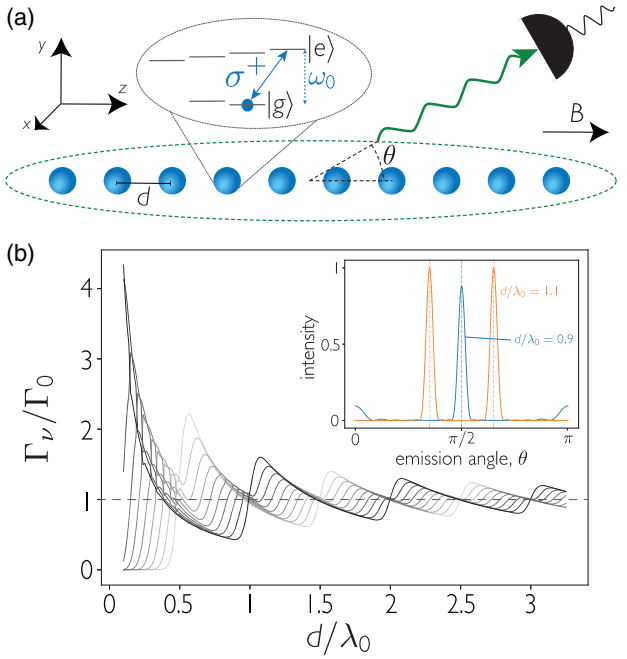

FIG. 1. A chain of excited atoms decays collectively, emitting correlated photons. (a) Schematic of the considered setup. Atoms have resonance frequency ω_0, and are separated by a constant distance d. The relevant transition is selected via a small magnetic field. (b) Decay rates of the jump operators $\{\hat{\mathcal{O}}_\nu\}$ for $N = 10$ atoms. Each operator approximates a spin wave with wave vector k_ν, with the darkest (lightest) lines corresponding to spin waves with minimum (maximum) wave vector. Inset: Angular emission pattern (in arbitrary units) following action of the most subradiant operator on a fully inverted array, measured by detectors of width $\Delta\theta = 0.01\pi$. Dashed lines are analytically obtained angles of peak emission, $\theta_{\max} = \arccos(\pm k_\nu/k_0)$ with $k_\nu = 0(\pi/d)$ for $d = 0.9(1.1)\lambda_0$.

transition of the ith atom at position $\mathbf{r}_i = \{x_i, y_i, z_i\}$. The coherent and dissipative interactions between atoms i and j are [38,39]

$$J^{ij} - i\frac{\Gamma^{ij}}{2} = -\frac{\mu_0\omega_0^2}{\hbar}\wp^* \cdot \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j, \omega_0) \cdot \wp, \tag{3}$$

where $\wp = (|\wp|/\sqrt{2})(\hat{x} + i\hat{y})$ is the dipole matrix element of the circularly polarized transition σ^+, and $\mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j, \omega_0)$ is the propagator of the electromagnetic field between positions $\mathbf{r}_i$ and $\mathbf{r}_j$ [9]. The scattered field along the axis of the chain is σ^+ polarized and the atoms behave as two-level systems even in the presence of complex hyperfine structure [40–42].

An ensemble of N atoms decays collectively, via a set of N jump operators, $\{\hat{\mathcal{O}}_\nu\}$, with associated decay rates $\{\Gamma_\nu\}$. These operators are eigenstates of the dissipative interaction matrix $\mathbf{\Gamma}$ with elements Γ^{ij} [13–15]. The master equation can be written in terms of these operators as

$$\dot{\rho} = -\frac{i}{\hbar}[\mathcal{H}, \rho] + \sum_{\nu=1}^{N}\frac{\Gamma_\nu}{2}(2\hat{\mathcal{O}}_\nu\rho\hat{\mathcal{O}}_\nu^\dagger - \rho\hat{\mathcal{O}}_\nu^\dagger\hat{\mathcal{O}}_\nu - \hat{\mathcal{O}}_\nu^\dagger\hat{\mathcal{O}}_\nu\rho). \tag{4}$$

The operators' decay rates can be superradiant, i.e., $\Gamma_\nu > \Gamma_0$, or subradiant, i.e., $\Gamma_\nu < \Gamma_0$, with $\Gamma_0 \equiv \Gamma^{ii}$ the single-atom spontaneous emission rate.

Jumps happen stochastically, and operators act on all atoms with a set of amplitudes and phases sensitive to d and the atomic quantization axis. The states $\{\hat{\mathcal{O}}_\nu^\dagger|g\rangle^{\otimes N}\}$ ($\{\hat{\mathcal{O}}_\nu|e\rangle^{\otimes N}\}$) form an orthonormal basis for the single-excitation ("single-hole") system. The jump operators can be classified according to the symmetries of the Lindblad operator. For an infinite 1D array, these are discrete translations along $\hat{z}$. Thus, the operators correspond to Bloch waves with a wave vector along the chain direction k_ν, i.e., $\hat{\mathcal{O}}_\nu = (1/\sqrt{N})\sum_{i=1}^{N} e^{-ik_\nu z_i}\hat{\sigma}_{ge}^i$. In 1D geometries, the collective decay rates Γ_ν change with d featuring sharp oscillations at $d \simeq n\lambda_0/2$ with $n \in \mathbb{N}$ [7,43,44], as shown in Fig. 1(b). For $d = n\lambda_0/2 + \epsilon$ ($d = n\lambda_0/2 - \epsilon$), with $\epsilon \to 0^+$, there are a small number of superradiant (subradiant) operators, with the majority of rates weakly subradiant (superradiant). These oscillations arise from 1D lattice sums and can be understood by analogy with the decay of a dipole in a cavity [7].

When a jump occurs, a photon is emitted. We calculate the emission angle of the radiated photons by means of directed-detection operators, following Carmichael and coworkers [13–15]. Photon detection at a point $\mathbf{R} = (r, \theta, \phi)$ corresponds to action of the operator

$$\hat{\mathcal{D}}(\theta, \phi) = \sqrt{\frac{3\Gamma_0}{8\pi}\left(1 - \frac{\sin^2\theta}{2}\right)d\Omega}\sum_{j=1}^{N} e^{-ik_0 z_j\cos\theta}\hat{\sigma}_{ge}^j, \tag{5}$$

where $d\Omega$ is a solid-angle differential. The detectors are assumed to be in far field, such that $|\mathbf{R}| \gg \lambda_0, Nd$. The probability of a photon detection in direction (θ, ϕ) is $P(\theta, \phi)d\Omega = \langle\hat{\mathcal{D}}^\dagger(\theta, \phi)\hat{\mathcal{D}}(\theta, \phi)\rangle$. A (square) photon detector of finite solid angle $\Delta\Omega$ and angular width $\Delta\theta$ sees intensity

$$\mathfrak{I}(\theta) = \frac{\Delta\Omega}{\Delta\theta}\int_{\theta-\Delta\theta/2}^{\theta+\Delta\theta/2} P(\theta')d\theta'. \tag{6}$$

Photon emission caused by action of a jump operator is directional. Figure 1(b) shows the angular distribution of a photon emitted during the action of the most subradiant operator on the fully inverted array. The maximal emission angles, calculated by considering correlations between jump and directed-detection operators, are [see Supplemental Material (SM) [45]]

$$\theta_{\max} = \arccos\left(\frac{n\lambda_0}{d} \pm \frac{k_\nu}{k_0}\right), \qquad n \in \mathbb{Z}. \tag{7}$$

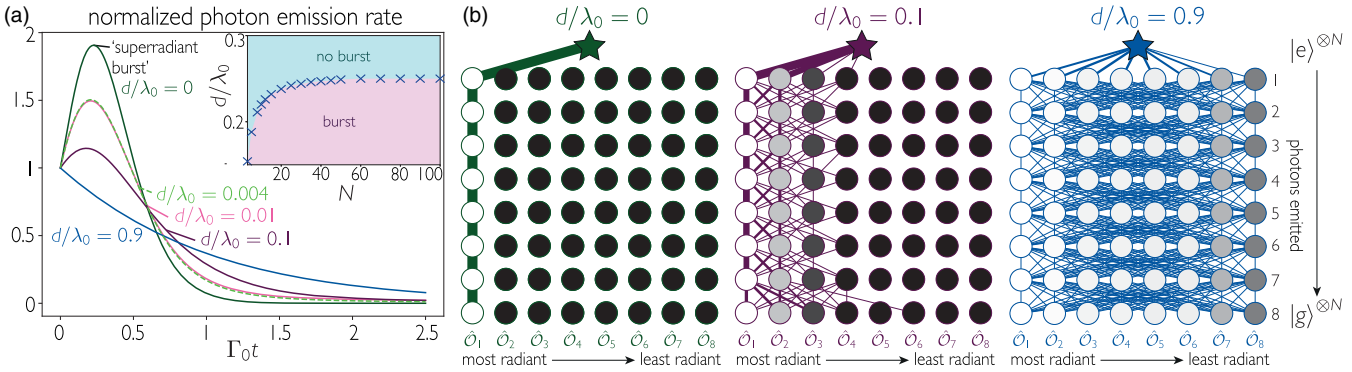

FIG. 2. Decay of a fully inverted chain and stochastic action of collective jump operators. (a) Photon emission rate normalized by atom number $[-d\langle n_{\rm exc}(t)\rangle/d(N\Gamma_0 t)]$ of an array of $N = 8$ atoms for different interatomic distances. As d is increased, the emission rate at early times shows a transition from an increase (superradiant burst) to a decrease. Inset: Boundary between regions. We estimate a burst occurs if the emission rate is larger at $N\Gamma_0 t = 10^{-4}$ than at $t = 0$. (b) Illustrations of jump operator action during the decay, for different lattice constants. The star represents the initial state $|e\rangle^{\otimes N}$. Circles represent action of one of the N different jump operators $\hat{\mathcal{O}}_\nu$, colored and displayed in order from most superradiant (white) on the left to most subradiant (black) on the right. Line thickness represents the likelihood of a particular path, based on a set of 1000 trajectories. For $d = 0.1\lambda_0$, some trajectories are extremely subradiant (23 trajectories not fully deexcited by $\Gamma_0 t = 500$ are omitted).

Here, the $\pm$ accounts for the mirror reflection symmetry of a finite chain (whose jump operators carry $\pm k_\nu$ wave-vector components). Jump operators cannot be expressed in terms of directed-detection operators [14]. For multiple holes and excitations, the intensity pattern may contain additional lobes due to atomic correlations.

A fully inverted array develops correlations as it decays. The rate of change of the atomic population, $\langle \hat{n}_{\rm exc}\rangle = \sum_{i=1}^{N}\langle \hat{\sigma}^i_{ee}\rangle$, dictates the photon emission rate

$$R(t) = -\frac{d\langle \hat{n}_{\rm exc}(t)\rangle}{dt} = \sum_{\nu=1}^{N}\Gamma_\nu\langle \hat{\mathcal{O}}_\nu^\dagger\hat{\mathcal{O}}_\nu\rangle. \tag{8}$$

At $t = 0$, all atoms are excited, and there are not any correlations between them ($\langle \hat{\mathcal{O}}_\nu^\dagger\hat{\mathcal{O}}_\nu\rangle = 1\,\forall\,\nu$). By definition, $\sum_\nu \Gamma_\nu = \mathrm{Tr}\,\mathbf{\Gamma} = N\Gamma_0$, and $R(t=0) = N\Gamma_0$. Since the atoms are uncorrelated, the initial decay is the sum of N independently decaying atoms.

Dicke superradiance ($d = 0$) is a unique situation, as there is only one jump operator with nonzero decay rate, $\hat{\mathcal{O}}_D$. That operator has rate $N\Gamma_0$ and acts identically on all atoms, i.e., $\hat{\mathcal{O}}_D = (1/\sqrt{N})\sum_{i=1}^{N}\hat{\sigma}^i_{ge}$. The decay rate $R_D(t) = N\Gamma_0\langle \hat{\mathcal{O}}_D^\dagger\hat{\mathcal{O}}_D\rangle$ is maximized with half the atoms excited. This gives rise to the superradiant burst seen in Fig. 2(a), where the peak rate of photon emission scales as N^2 and occurs at some finite time [2]. Since there is a single jump operator, decay is never subradiant. Dicke superradiance seems not to be recovered as $d \to 0$ [see Fig. 2(a) and the SM [45]]: the emission rate saturates to a different asymptotic curve, suggesting that Dicke superradiance is not analytically connected to this regime. Rings show similar behavior [45]. We attribute this saturation to a complex interplay of stimulation and competition between the set of jump operators that deexcites the array, as discussed below.

In extended arrays, jump operators enhance their own action [1,13,14] but compete with each other. This occurs due to correlations induced by photon emission, irrespective of whether the photon is detected or not. For the fully inverted array, the normalized probability of two different successive jumps ($\hat{\mathcal{O}}_\nu$ and $\hat{\mathcal{O}}_\mu$) can be calculated analytically. In the large N limit, it yields [45]

$$\tilde{g}^{(2)}(\tau = 0)|_{\nu,\mu} = \frac{\langle \hat{\mathcal{O}}_\nu^\dagger\hat{\mathcal{O}}_\mu^\dagger\hat{\mathcal{O}}_\mu\hat{\mathcal{O}}_\nu\rangle}{\langle \hat{\mathcal{O}}_\nu^\dagger\hat{\mathcal{O}}_\nu\rangle\langle \hat{\mathcal{O}}_\mu^\dagger\hat{\mathcal{O}}_\mu\rangle} \simeq 1 + \delta_{\nu\mu} - \frac{2}{N}. \tag{9}$$

While this is a process of spontaneous emission, each jump operator enhances itself (but not others), and thus, an effective stimulated emission of radiation occurs at certain angles, as jump operators are directional. The last term in the equation is a fermionic correction that illustrates that a single atom can only host a single excitation (there is a $1/N$ probability for two excitations to overlap in a chain of N atoms, and the factor of 2 arises because there are two identical ways to assign two excitations). For $N \geq 4$, $\tilde{g}^{(2)}(\tau = 0)|_{\nu,\nu} > 1\,\forall\,\nu$, i.e., even in very short chains, all operators enhance their own action.

The competition between different jump operators causes dephasing of the atomic state, reducing and eventually blocking superradiance. Following this argument, atoms in other geometries must also dephase, since for $d \neq 0$, there are always multiple operators and, thus, competition between decay paths. The set of likely deexcitation paths diversifies as d increases, as shown in Fig. 2(b), leading to faster dephasing. This reduces the intensity of the superradiant burst and brings it forward in time—no longer happening when half of the atoms are

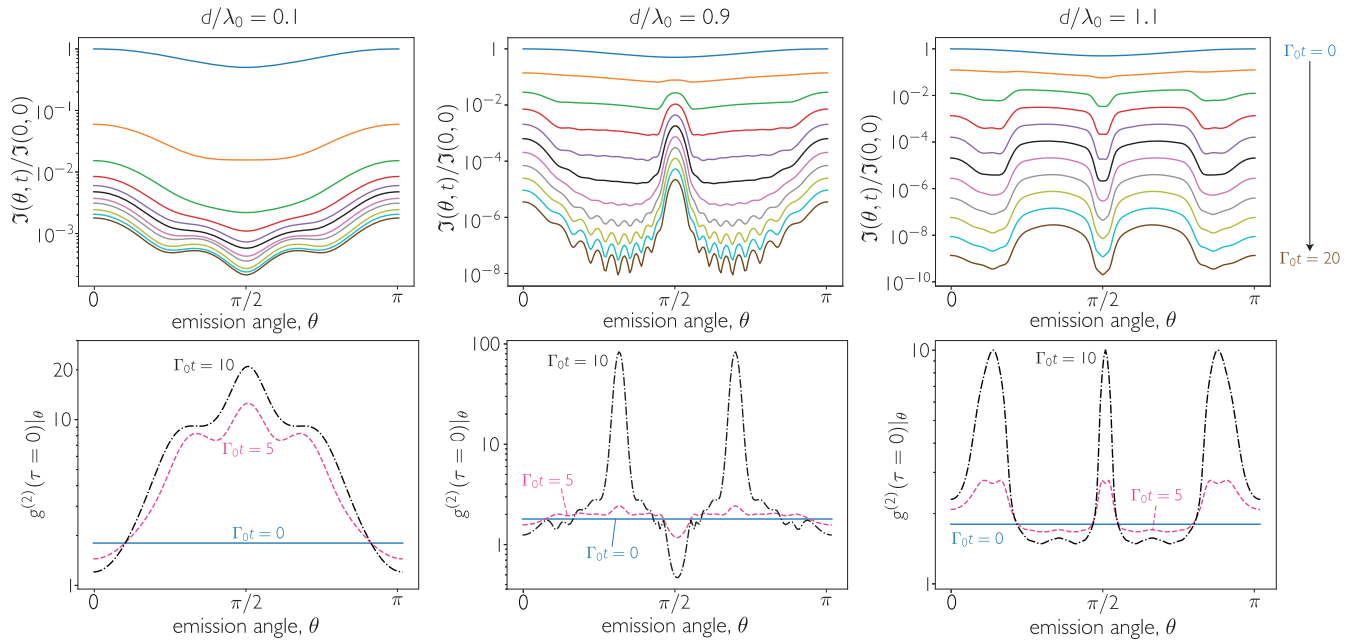

FIG. 3. Directional photon emission from a fully inverted chain of $N = 10$ atoms. (top) Intensity, normalized by value at $t = 0$, at far-field detectors of angular width $\Delta\theta = 0.01\pi$. Curves represent evenly spaced snapshots of the intensity profile for $\Gamma_0 t \in [0, 20]$. (bottom) Directional two-photon correlation function, $g^{(2)}(\tau = 0)|_{\theta,\theta}$ as defined in Eq. (10).

excited, but earlier—until the burst disappears. Each path has a likelihood dictated by the operator decay rates and the correlations induced as the ensemble deexcites. The only forbidden paths are those where the cumulative effect of the jump operators breaks the mirror symmetry about the center of the array.

We find that the superradiant burst survives at small enough interatomic distances despite being suppressed, as shown in Fig. 2(a). While the peak intensity is fainter and does not scale as N^2 [45], the photon emission rate initially increases. In the inset, we show the distance for which the superradiant burst disappears. We estimate that a superradiant burst will occur if the derivative of the photon emission rate at $t = 0$ is positive. We perform calculations for large atom numbers by truncating the Hilbert space to subspaces with up to two atoms in the ground state (i.e., maximum of two photons emitted). This captures well the early dynamics. As shown in the inset of Fig. 2(a), for long chains, superradiant features are retained for $d \lesssim 0.25\lambda_0$. At this upper limit, competition between jump operators becomes so strong that the burst becomes marginal and is eventually suppressed.

The action of different jump operators throughout the evolution leads to changes in the directionality of photon emission at different times, as shown in Fig. 3. The fully excited ensemble emits quite broadly in space. Without correlations, the atoms emit as independently radiating dipoles [13,14]. However, at late times, subradiant operators become dominant and emission is strongly peaked in a direction dictated by d. Angular emission is narrow for $d = 0.9\lambda_0$, as there is one dominant subradiant mode, but broad for $d = 1.1\lambda_0$ where multiple subradiant modes are important [see Fig. 1(b)]. Radiation in different directions is correlated [20]: emission at angle θ_1 enhances emission in directions that satisfy $\cos\theta_2 = \cos\theta_1 - n\lambda_0/d$, $n \in \mathbb{Z}$, as jump operator emission patterns are multilobed [45].

Self-enhancing (or stimulated) emission is confirmed by calculating the direction-dependent second order correlation function

$$g^{(2)}(\tau = 0)|_{\theta,\theta} = \frac{\langle \hat{\mathcal{D}}^\dagger(\theta)\hat{\mathcal{D}}^\dagger(\theta)\hat{\mathcal{D}}(\theta)\hat{\mathcal{D}}(\theta)\rangle}{\langle \hat{\mathcal{D}}^\dagger(\theta)\hat{\mathcal{D}}(\theta)\rangle^2}. \quad (10)$$

Figure 3 shows large, direction-dependent bunching in the field radiated by the array under evolution according to Eq. (4). At $t = 0$, $g^{(2)}(\tau = 0)|_{\theta,\theta}$ can be calculated analytically, yielding a spatially uniform value of $2 - 2/N$ [45], reproducing Dicke's result [1,15]. At late times, there are large peaks at intensity minima. While single photon emission is very unlikely, conditioned on one photon, a second is significantly more likely, such that pairs are relatively enhanced [17,19]. At late times, the signal can be sub-Poissonian in the direction of peak intensity [see plot for $d = 0.9\lambda_0$ in Fig. 3], as subradiance is predominantly a single-excitation effect and photon pairs are suppressed. Evidence of such directional statistics has been observed for two emitters [51].

Signatures of collective decay can be observed without fully inverting the array, but preparing spin coherent states, instead [52],

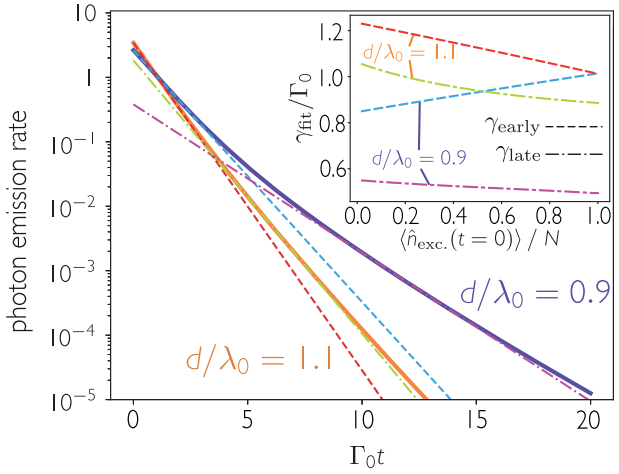

FIG. 4. Role of initial number of excitations on photon emission rate from spin coherent states. Photon emission (solid lines) from $|\varphi = 0.3, \mathbf{k} = k_0\hat{z}\rangle$ with average excitation number $\langle \hat{n}_{\rm exc}\rangle = \varphi N$ [see Eq. (11)]. Dashed lines show exponential fits, from which one extracts decay rate coefficients $\gamma_{\rm early}$ (dashed) and $\gamma_{\rm late}$ (dot-dashed). Inset: Early and late fitted decay rates as a function of the initial number of excitations. For all plots, $N = 10$. For $d = 0.9\lambda_0$, $\gamma_{\rm early}$ is fitted for $\Gamma_0 t \in [0, 4]$ and $\gamma_{\rm late}$ for $\Gamma_0 t \in [10, 20]$. For $d = 1.1\lambda_0$, $\gamma_{\rm early}$ is fitted for $\Gamma_0 t \in [0, 2]$ and $\gamma_{\rm late}$ for $\Gamma_0 t \in [5, 10]$.

$$|\varphi, \mathbf{k}\rangle = \prod_{i=1}^{N} \sqrt{1-\varphi}\,|g_i\rangle + e^{i\mathbf{k}\cdot\mathbf{r}_i}\sqrt{\varphi}|e_i\rangle, \qquad (11)$$

where $0 \le \varphi \le 1$. These states have a binomially distributed excitation number defined by probability φ, and an excitation expectation value $\langle \hat{n}_{\rm exc}\rangle = \varphi N$. Coherent spin states can be prepared experimentally by exciting the array with an intense pulse of duration $\tau \ll (N\Gamma_0)^{-1}, (J^{12})^{-1}$, to prevent collective effects.

Coherent spin states exhibit nonexponential temporal decay due to the interplay of multiple jump operators. The subradiant tail survives at distances accessible with current experimental capabilities, as shown in Fig. 4. This can be characterized by separately fitting the early and late dynamics, as demonstrated in experiments with atomic clouds in free space [53,54] and near a nanofiber [55]. The fitted decay rates of early dynamics depend on the initial atomic state; the decay is a many-body problem dependent on the density of excitations. Fits at late times do not depend so markedly on the number of excitations, since late dynamics are predominantly a single-excitation phenomenon independent of initial conditions. For large numbers of excitations, $\gamma_{\rm early}$ converges for all geometries as the fully excited state is uncorrelated. The contrast between early and late fits is significantly larger for $d = n\lambda_0/2 - \epsilon$, due to the differences in decay rates across each resonance. As shown in the SM [45], the drive can be used to imprint correlations on the array, some of which survive at long times and impact the radiation pattern.

Experimental realizations in the regime $d \sim \lambda_0$ should be feasible with current technologies. However, reaching the superradiant regime requires shorter interparticle distances. The limit of $d \lesssim 0.25\lambda_0$ established here could be satisfied with long-wavelength transitions (such that $\lambda_0 > 800$ nm) of lanthanide atoms trapped in optical lattices with wavelength near their strong transitions ~400 nm [56,57]. Even shorter interatomic distances can be reached in disordered ensembles [58,59], which constitute an interesting prospect for future work. Arrays of solid state emitters, such as localized excitonic quantum dots or strain-generated defects in 2D materials [60,61], are an additional playground for collective decay.

In conclusion, we have studied the collective decay of ordered chains of atoms in free space. We have found that superradiance survives significant interatomic separations, though Dicke's perfectly symmetric decay is lost at any finite distance in all ordered geometries. For separations comparable to the resonance wavelength, strong signatures of collective decay remain, and photon emission has directional features and a subradiant tail. These phenomena are robust to realistic experimental imperfections, namely, finite filling fraction and classical position noise [45]. Geometry can be used to control the many-body optical response of arrays. With conditional feedback control [62] (assisted by directional detection), it may pave the way toward the preparation of target entangled states, such as metrologically useful subradiant states.

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