

Emergence of Second-Order Coherence in Superfluorescence

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We experimentally investigate the second-order quantum coherence function of a superradiant burst in a cascaded quantum system. We chirally (i.e., direction dependently) couple about 900 cesium atoms to the forward-propagating mode of an optical nanofiber. We then prepare the ensemble close to the maximally excited state, where the subsequent collective emission of a burst is known as superfluorescence. Here, we observe that second-order coherence emerges in the course of the decay. This is a clear signature of the underlying collective dynamics that is also at the origin of the superradiant burst itself. We furthermore study the dynamics of the second-order coherence function of the emission in relation to the initial average dipole moment of the ensemble. In addition, by investigating the correlation of early and late photon emission events, we obtain evidence for fundamental shot-to-shot fluctuations in the delay of the emission of the burst. Our findings thus reveal surprising similarities between superradiance in cascaded and symmetrically coupled systems, despite the fact that they are described by fundamentally different coupling Hamiltonians.

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Introduction—The collective emission of radiation is an ubiquitous physical process that underlies devices such as lasers and phased-array antennas, applications such as optical quantum memories, and even phenomena in space such as astrophysical cyclotron masers [1–3]. Already in 1954, Dicke studied the collective emission originating from a dense ensemble of two-level quantum emitters prepared in the maximally excited state [4–7]. In this system, superfluorescence occurs, where the radiated optical power first increases with time, then reaches a maximum and, eventually, decays. Such a superradiant burst of light is a hallmark effect in many-body quantum optics and qualitatively differs from the exponential decay of the emission from independent atoms. Recently, it was theoretically [8] and experimentally [9] shown that a burst also occurs for a cascaded interaction between emitters. In this setting, an atom i influences the decay of atoms $j > i$ [10,11], but not vice versa as in the Dicke case. Cascaded interactions between quantum emitters can be engineered, for example with Rydberg atoms [12], acousto-optic control techniques [13], spin-orbit coupled Bose-Einstein condensates [14], and occur naturally for quantum emitters in optical near fields due to chiral light-matter interaction [15]. While superfluorescence and in general superradiant phenomena have been studied extensively [7,16–27],

important properties such as the coherence and the photon statistics of the burst emission have only been explored sparsely in the literature [9,28–31].

Here, we experimentally study the two-time second-order quantum coherence function [32], $g^{(2)}(t_1, t_2)$, of the superradiant burst emitted by a cascaded quantum system. Our system is realized with about 900 cesium atoms that are chirally coupled to the guided mode of an optical nanofiber. From these second-order correlations, we discern the regimes of superfluorescence and superradiance, directly observe the spontaneous build-up of second-order coherence of the radiation emitted by an ensemble that initially shows $g^{(2)}(0, 0) \approx 2$, and infer shot-to-shot fluctuations of the delay between the excitation and the burst emission. We compare our data to a stochastic simulation based on the truncated Wigner approximation, to the symmetric Dicke model, as well as to numerical calculations based on a cascaded master equation.

Setup and measurement—Our experimental setup is sketched in Fig. 1. We realize an optical trapping potential in the evanescent field surrounding the nanofiber-waist of a tapered optical fiber (TOF), by sending running wave blue-detuned laser light (wavelength 760 nm, power 20.5 mW) and forward- and backward-propagating red-detuned laser light (wavelength 1064 nm, powers 1.3 and 1.1 mW, respectively) through the nanofiber. This creates two diametral arrays of trapping sites along the nanofiber [33], which we probabilistically load with cold cesium atoms by overlapping the nanofiber with a molasses-cooled cloud of atoms from a magneto-optical trap (MOT). Because of the collisional blockade effect, each trapping site is filled with at most a single atom [34]. We prepare the

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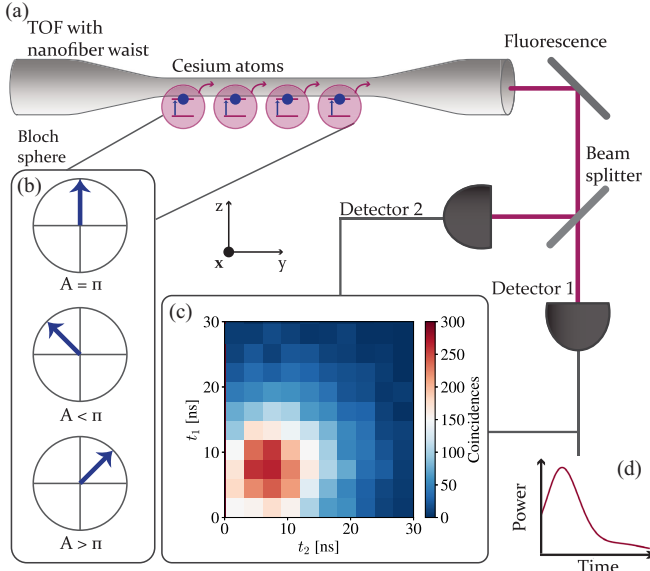


FIG. 1. (a) Schematic of the experimental setup. An ensemble of about 900 cesium atoms (red circles) are optically trapped near the surface of an optical nanofiber. A pulse of nanofiber-guided resonant light coherently excites the ensemble close to the maximally excited state, which predominantly reemits the light into the forward-propagating nanofiber mode, thereby realizing a cascaded quantum system. The optical power and its second-order correlation are obtained using a Hanbury Brown-Twiss detection setup. (b) Representation of the initial state on the Bloch sphere, parametrized by the Rabi pulse area A . (c) Measured two-photon coincidences (bin size: 3 ns). A maximum of roughly 250 two-photon coincidences occurs at $t_1 = t_2 \approx 7.5$ ns for 10^7 excitation cycles. (d) Sketch of the optical power.

atoms on only one side of the fiber in their motional ground state using side-selective degenerate Raman cooling (DRC) for 50 ms [35]. With this, we end up with a single one-dimensional array of trapped cesium atoms, which lies in the x - y plane, see Fig. 1(a). After the DRC, the internal state of the atoms is $|g\rangle = |6S_{1/2}, F=4, m_F=-4\rangle$, where the quantization axis is chosen along the z direction. We control the number of atoms, N , via the MOT loading time and determine it by measuring the optical depth (OD) with transmission spectroscopy through the TOF. All the measurements shown here are performed at an OD of about 40 ($N \approx 900$) at the start of the probing sequence [33].

Because of spin-momentum locking [36], the polarization of the waveguide mode at the position of the atoms is linked to its sense of propagation, forward or backward. In particular, for a single atom, the radiation emitted on the σ^- -polarized D2-cycling transition $|g\rangle \rightarrow |e\rangle = |6P_{3/2}, F=5, m_F=-5\rangle$ is coupled to the forward-propagating nanofiber mode with an efficiency $\beta = \Gamma_{\text{fwd}}/\Gamma_{\text{total}} = 0.01$, where Γ_{fwd} is the rate of photon emission into the forward-propagating nanofiber-guided mode and Γ_{total} is the rate of photon emission into all available

optical modes. The coupling to the backward-propagating mode is about ten times smaller.

Thanks to this so-called chiral coupling, the nanofiber-coupled atoms thus effectively realize a cascaded quantum system [10,11,15]. We coherently drive the $|g\rangle \rightarrow |e\rangle$ transition with a fiber-guided forward-propagating resonant optical Rabi pulse of duration $T_{\text{pulse}} = 4$ ns, which is shorter than the excited-state lifetime of $\tau = 30.5$ ns [37]. In a simplified picture this prepares the atoms close to the product state

$$|\psi_0\rangle = \bigotimes_{k=1}^N \left[\cos\left(\frac{A}{2}\right) |g_k\rangle - i \sin\left(\frac{A}{2}\right) |e_k\rangle \right], \quad (1)$$

which is depicted on the Bloch sphere in Fig. 1(b). Here, the atomic index $k = 1, \dots, N$ increases along the pulse propagation direction. The Rabi pulse area $A = \Omega T_{\text{pulse}}$ is defined by the Rabi frequency Ω applied to the first atom. In practice, because of attenuation of the pulse upon propagation through the atomic ensemble and due to inhomogeneous atom-waveguide coupling, the atoms experience slightly different pulse areas $A_k \approx A$, see discussions in Refs. [9,37]. In addition, the atoms are not exactly prepared in a product state. We will comment on this later in the Letter, when the effects of this small imperfection become apparent.

In each experimental run, we repeatedly excite the ensemble 400 times within 80 ms. We counteract the heating that results from this resonant probing by continuously cooling the ensemble by applying DRC on the D1 line ($|6S_{1/2}, F=4\rangle \rightarrow |6P_{1/2}, F=4\rangle$) with a free-space laser, achieving a survival of at least 75% of the atoms by the end of the probing period. Because the scattering rate of the D1 light is sufficiently small, this cooling does not alter the burst dynamics. We verified that this decrease of the atom number has only a marginal influence on the data; a detailed analysis can be found in Supplemental Material [38].

A fully inverted and sufficiently large ensemble (in our case $N \gg 100$ and $A \approx \pi$) then radiates a superradiant burst as sketched in Fig. 1(d) and experimentally investigated in Ref. [9]. A hybrid photodetector with large dynamic range (Hamamatsu, R10467, dead time < 2 ns) captures both this superradiant burst and the preceding transmitted excitation pulse. For the two-time correlation measurement of the burst, we split the light into two equal parts and delay the second half by about 100 ns using a 20-m-long optical fiber before it reaches the same detector as the earlier first half. Since the decay dynamics is much faster than 100 ns, this allows us to measure the two-photon coincidence rate, $n_c(t_1, t_2)$. Here, t_1 and t_2 are measured from the end of the Rabi pulse. A measurement of two-photon coincidences during a superradiant burst from an ensemble that is initially prepared close to the maximally excited state is shown in Fig. 1(c). The normalized second-order coherence function is then given by

$$g^{(2)}(t_1, t_2) = \frac{n_c(t_1, t_2)}{n_1(t_1)n_2(t_2)}, \quad (2)$$

where $n_j(t_j)$ is the photon detection rate on detector j at time t_j with $j = 1, 2$. Note that n_c , n_1 , and n_2 are averaged over many realizations and $g^{(2)}(t_1, t_2)$ is computed from these averages.

Initial second-order coherence function—Let us first discuss the value of the second-order coherence function of the light that is emitted right after the excitation pulse, $g^{(2)}(0, 0)$, which is shown in Fig. 2(a) as a function of A (black circles). The data reaches a peak value of $g^{(2)}(0, 0) = 2$ for $A \approx 1.1\pi$, falls off over a range of about $\pm 0.1\pi$, and decreases to $g^{(2)}(0, 0) \approx 1$ for smaller or larger pulse areas. The peak position can be understood in the following way. Because of the absorption of the pulse along the ensemble, a pulse area at the first atom slightly larger than π (here, $A \approx 1.07\pi$) maximizes the mean excitation stored in the ensemble [37]. For this case of maximum excitation, the initial state is $|\psi_0\rangle \approx \otimes_k |e_k\rangle$ and the ensemble-averaged dipole moment is close to zero, cf. Bloch sphere in Fig. 1(b).

In order to provide an intuitive understanding of our experimental observations, we first make the simplified assumption the atoms are prepared in $|\psi_0\rangle$ as defined in Eq. (1) and calculate $g^{(2)}(0, 0)$. The total electric field $\hat{E}$ in the detected mode is given by the sum over the contribution of all atomic dipoles $\hat{\sigma}_k = |g_k\rangle\langle e_k|$, i.e., $\hat{E} \propto \sum_k \hat{\sigma}_k$. As shown in Supplemental Material, we find the following expression

$$g^{(2)}(0, 0) = \frac{\langle \psi_0 | \hat{E}^\dagger \hat{E}^\dagger \hat{E} \hat{E} | \psi_0 \rangle}{\langle \psi_0 | \hat{E}^\dagger \hat{E} | \psi_0 \rangle^2} \approx 2 - \frac{1}{\left(1 + \frac{1}{N \cos^2(A/2)}\right)^2}, \quad (3)$$

where the right-hand side is exact in the large-ensemble limit of $N \gg 1$. We show this prediction as a gray line for $N = 900$ in Fig. 2(a). The width of the peak predicted for $g^{(2)}(0, 0)$ as a function of A is $\approx 2\pi/\sqrt{N}$. As stated above, the state in which we prepare the atoms deviates from a product state. Still, the width predicted by Eq. (3) agrees well with our experimental observation. We can also model the full dynamics including the excitation process by a Rabi pulse that propagates through the ensemble using a stochastic numerical model based on the truncated Wigner approximation for spins (TWA) [45,46], see below. The corresponding prediction is shown as the purple line in Fig. 2(a). The TWA-based model thus explains the shift of the peak by $\sim 0.07\pi$ and predicts a width in very good agreement with the experimental observations.

Dynamics of the second-order coherence function—In Figs. 2(b)–2(d), the blue shaded areas show the measured optical power, $P(t)$, which is averaged over more than 10^6 excitation pulses, as the blue shaded area, and the

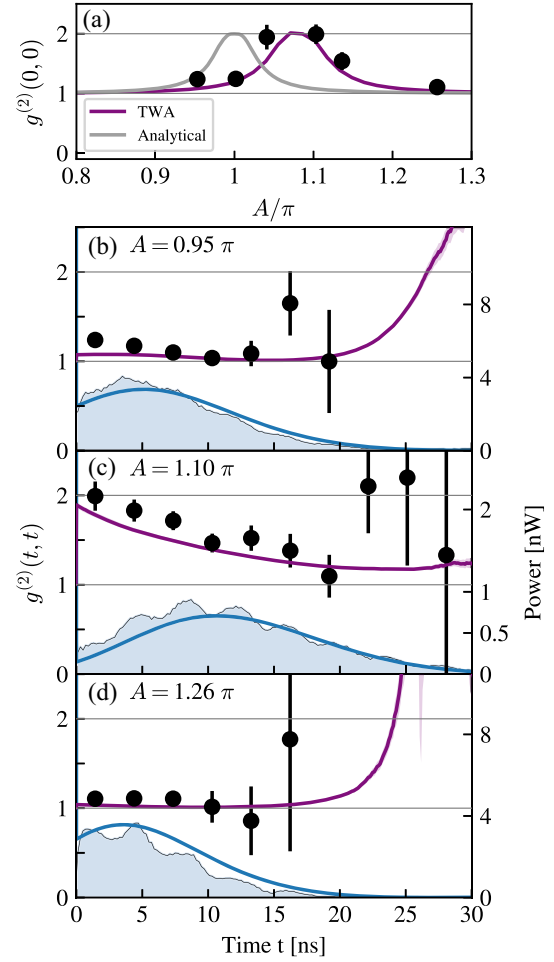


FIG. 2. (a) Second-order coherence function $g^{(2)}(0, 0)$ of the initial emission as a function of the excitation Rabi pulse area A (black circles with Poissonian error bars). The gray and purple theory lines are, respectively, an analytical prediction according to Eq. (3) and a numerical prediction of the TWA model for $N = 900$ atoms. (b)–(d) $g^{(2)}(t, t)$ for three values of A , as indicated. For $A = 1.10\pi$, $g^{(2)}(t, t)$ develops from 2 to 1 during the burst. For $A \neq 1.10\pi$, $g^{(2)}(t, t) = 1$ is constant. The purple lines show TWA predictions with the one-sigma error due to a finite number of computed trajectories (shaded areas). The TWA reproduces the data well, with a remaining discrepancy of roughly 10%. In (b)–(d) we omitted those data points, where we did not observe at least a single coincidence. The optical power $P(t)$ is shown as the blue area with the corresponding TWA prediction as the dark blue line. The oscillations in $P(t)$ are quantum beats that originate from weak excitation of the hyperfine state $|6P_{3/2}, F = 4\rangle$ (see Supplemental Material). The power at $t = 0$ in (c) exceeds the value expected from an ensemble in a product state, where the emission from the atoms is independent, see the main text and SM for explanation.

normalized second-order coherence function at equal times, $g^{(2)}(t, t)$, during the burst as black circles for three different initial atomic states characterized by the Rabi pulse area A . The characteristic feature of the superradiant decay of an

inverted atomic ensemble is the burst of the optical power [9]. It is most prominent in panel (c), where the initial state is close to maximal excitation. Here we measure a decrease of $g^{(2)}(t, t)$ during the burst emission from its initial value of $g^{(2)}(0, 0) \approx 2$ to $g^{(2)}(t, t) \approx 1$ at $t \approx 19$ ns. In stark contrast, we observe a constant $g^{(2)}(t, t) \approx 1$ for the entire duration of the burst emission in panels (b) and (d) of Fig. 2, where the ensemble's average initial dipole moment is nonzero.

The theoretical modeling of such second-order correlations for as many as one thousand atoms requires state-of-the-art approaches [8,47–55]. Here, we apply an efficient, recently developed simulation tool based on the TWA [45,56], which can describe many emitters and many excitations. In our model, the system dynamics are mapped to a set of $2N$ coupled, nonlinear stochastic differential equations, which we numerically solve for thousands of atoms, to predict both the output power (blue lines in Fig. 2) and $g^{(2)}(t, t)$ (purple lines). For the details on the implementation of the TWA, see Ref. [46]. Our modeling includes the coherent Rabi excitation process, where we account for the attenuation of the excitation pulse along the ensemble as well as for the inhomogeneous atom-waveguide coupling due to the atom's thermal motion. Let us note that the TWA describes both $g^{(2)}(t, t)$ and power, for a fixed set of system parameters, very well.

Before we discuss the dynamics of $g^{(2)}(t, t)$ for the case of the maximally excited ensemble in Fig. 2(c), we note that, at this pulse area, the measured initially emitted power of ~ 250 pW exceeds the expected value of $P(t=0) = 74$ pW for about 900 fully excited cesium atoms. The latter value is given by $N\Gamma_{\text{fwd}}\hbar\omega$, where ω is the angular frequency of the cesium D2 transition. This ~ 3.5 -fold elevated initial power stems from two contributions: First, the used excitation pulse area is too large by $\sim 2\%$, leading to a residual average dipole moment and a corresponding coherent emission. Second, even the power of the incoherent emission is higher than for independent atoms, because of correlations that build up during the excitation process. More details on both points are given in the SM.

In spite of the elevated initial power, which is partly due to residual coherent emission, we observe a $g^{(2)}(0, 0) \approx 2$. From the initial values of $g^{(2)}(0, 0) \approx 1$ in Figs. 2(b) and 2(d) we conclude that initial value of $g^{(2)}(0, 0) \approx 2$ in Fig. 2(c) is not due to technical fluctuations, but rather to the incoherent contribution to the initial emission. Thus, the initially emitted light field is not second-order coherent. However, it is partially coherent with the excitation light field. Quantitatively, at the working point in Fig. 2(c), we estimate that their first-order cross-correlation is reduced to $\sim 50\%$, see Supplemental Material. This places our measurement at the verge of superfluorescence [16,17].

Following the initial emission, second-order coherence builds up while the light is radiated. These characteristic

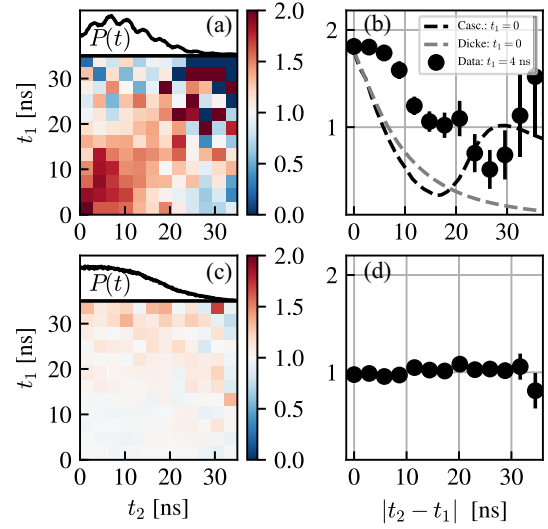


FIG. 3. (a) $g^{(2)}(t_1, t_2)$ of the superradiant burst at detection times t_1, t_2 . The trace of the average measured power (black line) is shown in the top panel. (b) Cut of $g^{(2)}(t_1, t_2)$ as a function of $|t_2 - t_1|$ (black circles with Poissonian error bars), reveals anticorrelations for delays > 20 ns. The gray (black) dashed line is a prediction based on the symmetric Dicke model (cascaded quantum system) for $\tilde{N} = 9$ and $\beta = 1$. (c),(d) Same analysis for a coherent laser pulse. Here, $g^{(2)}(t_1, t_2) = 1$ within the error bars.

dynamics of superfluorescence arise because the fluctuations of the atomic dipoles become correlated via the shared mode, and the Siegert relation [57] $g^{(2)}(t, t) = 1 + |g^{(1)}(t, t)|^2 = 2$ is increasingly violated [58]. To substantiate our interpretation that the emergence of second-order coherence is a collective phenomenon, we present a theoretical study of $g^{(2)}(t, t)$ as a function of the number of atoms N in the End Matter Section below. We note that the Siegert relation is also violated for an excitation Rabi pulse area sufficiently below and above maximal inversion [cf. Figs. 2(b) and 2(d)]. There, the correlations among the atoms do not spontaneously build up, but are already present at $t = 0$ as they are imprinted onto the atoms by the excitation pulse [9].

Shot-to-shot variations—Finally, we turn to the statistical properties of the light emitted from an ensemble prepared close to the maximally excited state by considering $g^{(2)}(t_1, t_2)$ for unequal times $t_1 \neq t_2$, depicted in Fig. 3(a). In Fig. 3(b), we show $g^{(2)}(t_{1,2} = 4 \text{ ns}, t_{2,1})$ as a function of the time difference $|t_2 - t_1|$. This data quantifies the correlation between the burst light at early and late times. Interestingly, the detection events are correlated, $g^{(2)}(t_1, t_2) > 1$, for small time differences and anti-correlated, $g^{(2)}(t_1, t_2) < 1$, for larger time differences. These anticorrelations are also apparent as blue regions in the bottom right and top left corner of Fig. 3(a). As a reference, we present the results of the same correlation measurement

performed by sending a coherent laser pulse of similar shape and duration in panels (c) and (d). Here $g^{(2)}(t_1, t_2)$ is constant and equals 1 within the error bars as expected given that there are no correlations between the detection of early and late laser photons. For the time being and to our knowledge, models based on the TWA cannot predict two-time correlators for 900 atoms. To compare our data to a theoretical prediction, we therefore employ two effective models with perfect atom-waveguide coupling ($\tilde{\beta} = 1$) and a much smaller atom number of $\tilde{N} = 9 \approx \beta N$, inspired by Refs. [7,26]. The first is the symmetric Dicke model [4], which can be solved efficiently, and the second is a cascaded quantum system [9–11,46], which we numerically solve with QuTiP (see Supplemental Material). The predictions are shown as the gray and black dashed lines in Fig. 3(b), respectively. We note that both models feature qualitatively similar transitions from initially $g^{(2)}(t_1, t_1) \approx 2$ to $g^{(2)}(t_1, t_2) < 1$ at time differences $|t_2 - t_1| < 15$ ns. However, at larger time differences both models exhibit qualitative differences, such as the increase of $g^{(2)}(t_1, t_2)$ for $|t_2 - t_1| > 15$ ns in the case of the cascaded interaction.

To understand these anticorrelations, let us assume that individual bursts have a duration τ_s , and that—despite the identical preparation of the atoms—each burst occurs after a varying delay with respect to the excitation pulse [7]. When averaging over many realizations, the resulting power trace will show a burst with a duration that is longer than τ_s . At the same time, the probability of the detection of two photons separated by more than τ_s is smaller in a single shot than on average. We thus interpret the observed anticorrelations of $g^{(2)}(t_1, t_2)$ as a signature of these fluctuations. For the case of symmetric coupling, such shot-to-shot variations of the burst are a known feature of superfluorescence and originate from the fact that the burst is triggered by a first spontaneous emission [20,60]. We note that the influence of experimental atom-number variations on the presented data is negligible and cannot explain the observed anticorrelations, see Supplemental Material.

Conclusion and outlook—In conclusion, we have measured and theoretically investigated the two-time second-order quantum coherence function, $g^{(2)}(t_1, t_2)$, of a superradiant burst emitted by a cascaded quantum system. This allowed us to observe the emergence of second-order coherence in superfluorescence. Remarkably, this occurs despite the absence of any feedback in a cascaded system.

In conjunction with the experimental results from Ref. [9], it becomes apparent that superradiance in a cascaded quantum system features many similarities with superradiance in the symmetric Dicke model despite the fundamentally different Hamiltonian, which does not feature a particle-exchange symmetry. For example, in both systems a superradiant burst appears, there are shot-to-shot fluctuations as a consequence of spontaneous

emission, and second-order coherence builds up. Regarding these three aspects, a recent experimental study with an elongated cloud of laser-trapped atoms, which feature symmetrical coupling in free space, shows similar results [61]. Still, our theoretical analysis also points to qualitative differences between both models, e.g., in the emitted power and the photon-photon correlations, at late times [46].

Further characterization of the photonic output state of cascaded quantum systems includes measurements of yet higher-order correlations, such as $g^{(3)}(t_1, t_2, t_3)$. Moreover, it would be interesting to investigate the emission of coherently and incoherently driven cascaded quantum systems in the steady-state. From a theoretical standpoint, such experiments allow one to benchmark state-of-the-art simulation methods of many-body quantum systems. From a conceptional point of view, they may shed light on the physics underlying superradiant lasing [18].

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End Matter

Build-up of second-order coherence as collective phenomenon—We study $g^{(2)}(t, t)$ as a function of the number of atoms N to substantiate our interpretation that the emergence of second-order coherence is a collective phenomenon. In a cascaded system with imperfect atom-mode coupling ($\beta < 1$), a burst only appears when N exceeds a threshold value, $N_{\text{thr}} = 1 + 1/\beta$, whereas the atoms decay independently for $N \ll N_{\text{thr}}$ [7,8]. In our case, N_{thr} is about 100. Because of low count rates, we cannot reliably measure $g^{(2)}(t, t)$ for small atom numbers in our experiment and therefore analyze this regime theoretically using the TWA model. In Fig. 4, we show the calculated $g^{(2)}(t, t)$ for an ensemble of atoms with a homogenous coupling strength of $\beta = 0.01$, which is initialized in $|\psi_0\rangle = \otimes_k |e_k\rangle$. Note that this simulation does not account for the effects of imperfect state preparation and inhomogeneous coupling, which are included in the simulations shown in Fig. 2 of the main Letter. The rate at which the second-order coherence builds up decreases when N is reduced and, importantly, for $N < N_{\text{thr}}$ (depicted in blue), $g^{(2)}(t, t)$ is a constant. In that regime, the ensemble remains in a product state $\hat{\rho}(t) = \otimes_k \hat{\rho}_k(t)$, where $\hat{\rho}_k(t)$ is the density matrix of the

k^{th} atom, yielding $g^{(2)}(t, t) = g^{(2)}(0, 0) = 2(1 - 1/N)$. These results, which are based on the TWA, further support the conclusion that a build-up of second-order coherence in our experiment is a signature of a departure of the ensemble from the product state, i.e., $\hat{\rho}(t) \neq \otimes_k \hat{\rho}_k(t)$.

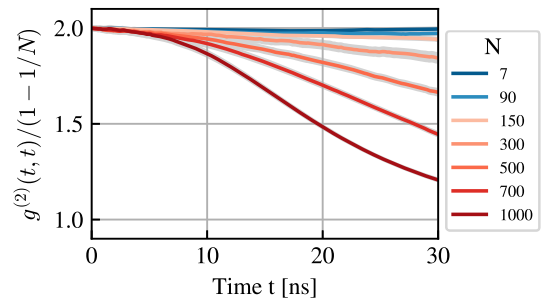


FIG. 4. TWA simulation of $g^{(2)}(t, t)$ for different numbers of atoms. For $N < N_{\text{thr}} \approx 100$ (blue) no burst occurs [9] and $g^{(2)}(t, t) = 2$ is constant. For $N > N_{\text{thr}}$ (red), $g^{(2)}(t, t)$ evolves toward 1, i.e., second-order coherence builds up during the collective emission.