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PAPER

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T C H Liew¹, Y G Rubo², A S Sheremet^{3,4}, S De Liberato⁵, I A Shelykh^{1,3,6}, F P Laussy^{4,7} and A V Kavokin^{4,5,8,9}¹ Division of Physics and Applied Physics, Nanyang Technological University, 637371, Singapore² Instituto de Energias Renovables, Universidad Nacional Autónoma de México, Temixco, Morelos, 62580, Mexico³ National Research University for Information Technology, Mechanics and Optics (ITMO), 197101 St.-Petersburg, Russia⁴ Russian Quantum Center, Novaya 100, 143025 Skolkovo, Moscow Region, Russia⁵ School of Physics and Astronomy, University of Southampton, Highfield, Southampton, SO17 1BJ, UK⁶ Science Institute, University of Iceland, Dunhagi-3, IS-107, Reykjavik, Iceland⁷ Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, E-28049, Spain⁸ Spin Optics Laboratory, St.-Petersburg State University, 198504 Peterhof, St.-Petersburg, Russia⁹ CNR-SPIN, Viale del Politecnico 1, Rome I-00133, ItalyE-mail: tchliw@gmail.com**Keywords:** bosonic quantum cascade lasers, superbunching, quantum well excitons, second order coherence, coherence in low dimensional systems**Abstract**

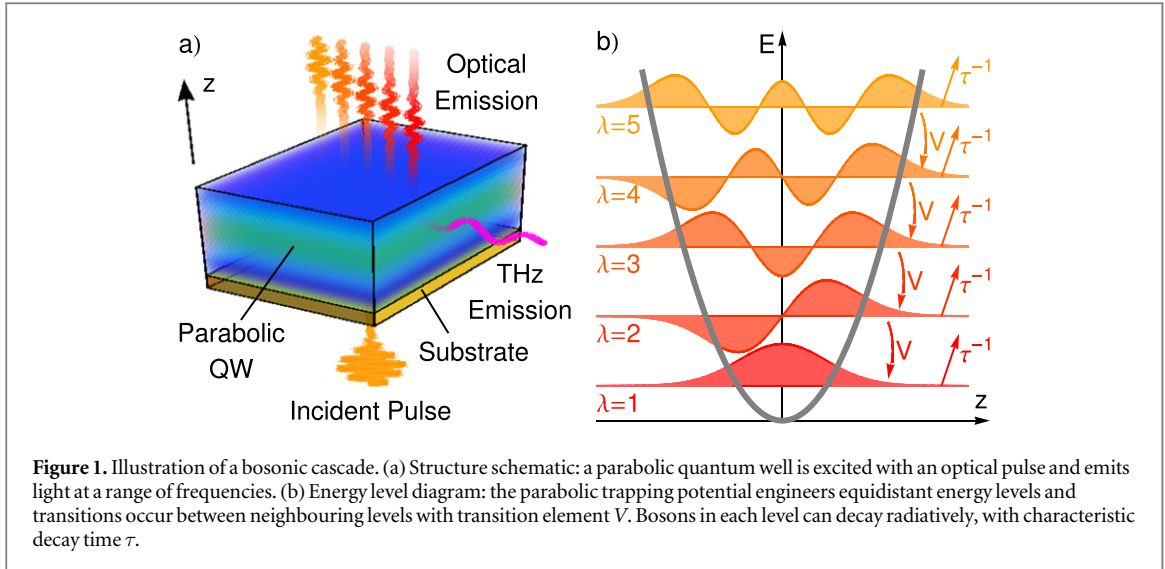
Bosonic cascades formed by lattices of equidistant energy levels sustaining radiative transitions between nearest layers represent a unique system to study correlated optical phenomena. We show how the light emitted by condensates in the visible range introduces a new regime of emission for cascade systems. Namely, the quantum statistics of bosonic cascades exhibits superbunching plateaus. This demonstrates further potentialities of bosonic cascade lasers for the engineering of correlated properties of light useful for imaging applications.

1. Introduction

A dissipative bosonic level coupled to a reservoir of higher levels may exhibit non-equilibrium condensation above a threshold density, where the bosonic stimulation of relaxation processes can cause the accumulation of particles in the lowest energy state [1–4]. In solid-state systems, this effect has resulted in the development of exciton-polariton lasers in a range of inorganic [5–8] and organic [9] based systems. The quantum optical properties of polariton lasers are well studied, where they are typically characterized by a second order coherence of the lasing mode close to unity [10–12]. The reservoir of higher energy modes is generally expected to remain in an incoherent/chaotic state, where each mode is only weakly occupied.

Bosonic cascade lasers are a recently proposed variant of polariton lasers in which relaxation takes place down a successive chain of bosonic levels that are equally and well-separated in energy [13]. They are different to fermionic cascade lasers [14], as the strength of relaxation processes is enhanced by bosonic stimulation [15]. Unlike conventional polariton lasers, the quantum statistics of bosonic cascades has yet to be explored.

In this work, we open a new dimension for bosonic cascades by showing that their emission produces a marked superbunching, thanks to their specificity of making coexist multiple macroscopic coherent states. We consider a system of exciton (exciton-polariton) bosonic condensates trapped in a parabolic potential that provides equidistant energy spacing between quantum confined exciton (exciton-polariton) states [16]. We allow for the cascade relaxation in this system: the excitons can transfer from the top to the bottom of the cascade sequentially passing all the energy levels. Apart from the cascade relaxation, excitons can decay radiatively emitting photons. We study the statistics of photons emitted from each exciton level as a function of time assuming that the system is excited at one of the high levels with a coherent light pulse. Unlike [13, 15] we do not consider the terahertz lasing that can be produced by the cascade relaxation process in the presence of an external terahertz cavity. There is no stimulation of the cascade process by the terahertz mode and, crucially for our effect, there is no absorption of terahertz photons by the cascade, allowing relaxation going down the ladder only.



The quantum description of superbunching in the presence of macroscopically occupied states is a complex problem which we can tackle here with stochastic quantum Langevin equations. This description of bosonic cascades' statistical properties confirms their departures from other types of lasing [17]. Unlike other proposals for giant superbunching, we do not require strong nonlinear interactions at the single particle level [18], the engineering of feedback [19], or large cooperativity as in fermionic realizations [20]. The mechanism allows in particular to involve a large number of particles, and could be of interest in ghost imaging applications [21, 22] or random number generation [23, 24].

2. Theory

We consider a bosonic cascade with N energy levels labelled from $\lambda = 1$ to N , as illustrated in figure 1. A parabolic quantum well engineers a set of bosonic levels equidistant in energy [13]. We account for the dominant transitions which are between neighbouring excitonic levels $\lambda + 1$ and λ , and we account for the radiative decay of all levels at a rate τ^{-1} . The transition rates between levels are stimulated by the bosonic nature of the particles. The quantum state of the system is described by the density matrix $\hat{\rho}$, which evolves according to the Liouville equation:

$$\begin{aligned} \frac{d\hat{\rho}}{dt} = & \frac{1}{2\tau} \sum_{\lambda=1}^N ([\hat{a}_\lambda, \hat{\rho}\hat{a}_\lambda^\dagger] + [\hat{a}_\lambda \hat{\rho}, \hat{a}_\lambda^\dagger]) \\ & + \frac{V}{2} \sum_{\lambda=1}^{N-1} ([\hat{a}_\lambda^\dagger \hat{a}_{\lambda+1}, \hat{\rho}\hat{a}_{\lambda+1}^\dagger \hat{a}_\lambda] + [\hat{a}_\lambda^\dagger \hat{a}_{\lambda+1} \hat{\rho}, \hat{a}_{\lambda+1}^\dagger \hat{a}_\lambda]), \end{aligned} \quad (1)$$

where \hat{a}_λ annihilates a boson from level λ and V is the scattering rate. Since the transitions between levels are incoherent and independent of phase correlations, the Hamiltonian evolution of each level is composed of a pure phase evolution of each level, which can be absorbed into the definition of the field operators via unitary transformation. We do not consider the spatial degree of freedom of excitons in the 2D plane in our theory, as it would make the corresponding Hilbert space too large to study. While formally our theory is then representative of a 'parabolic dot', we still expect it to have relevance to a 2D system in which the parabolic quantum well would form a series of subbands. In the limit of zero acoustic phonon temperature, only the zero in-plane wavevector state of each subband would be macroscopically occupied. Any excitons undergoing spontaneous scattering processes to non-zero wavevector states would then be returned to the zero in-plane wavevector states by stimulated scattering. Intuitively, the overall effect of the additional degrees of freedom would then be a renormalization of the lifetime τ and possible dephasing (which we will consider in appendix B).

Dotting equation (1) with Fock states and neglecting correlations between levels in the Born-Markov approximation leads to the Boltzmann equations, which describe well the average populations of each state and thereby provide the mean-field theory of bosonic cascades [13]. To go beyond this semi-classical description, and in particular to compute the photon statistics, correlations between states must be retained. A brute-force approach, however, is restricted to few levels and each with a modest occupancy. To access the most general cases of bosonic cascades, with macroscopic occupation of the states, we therefore expand the density matrix on the natural basis for this problem, which is that of coherent states [25]:

$$\hat{\rho} = \int \mathcal{P}(\alpha_1, \beta_1, \dots, \alpha_N, \beta_N) \hat{\Lambda}(\alpha_1, \beta_1, \dots, \alpha_N, \beta_N) d\mu, \quad (2)$$

where:

$$\hat{\Lambda}(\alpha_1, \beta_1, \dots, \alpha_N, \beta_N) = \frac{|\alpha_1, \dots, \alpha_N\rangle \langle \beta_1^*, \dots, \beta_N^*|}{\langle \beta_1^*, \dots, \beta_N^* | \alpha_1, \dots, \alpha_N \rangle}, \quad (3)$$

with \mathcal{P} the positive- P distribution, which differs from the Glauber-Sudarshan distribution in allowing for non-diagonal coherent state projectors. The complex numbers α_λ and β_λ are independent variables covering the whole complex plane. The integration measure is $d\mu = d^2\alpha_1 d^2\beta_1 \dots d^2\alpha_N d^2\beta_N$ and for ease of notation we write $\vec{\alpha} = (\alpha_1, \beta_1, \dots, \alpha_N, \beta_N)$. $\vec{\alpha}_n$ will be used to refer to an element of this vector (of length $2N$), while the notations α_λ and β_λ will still be used where λ is the level index. Writing the density matrix in terms of the positive- P distribution (equation (2)) transforms the master equation equation (1) into a Fokker-Planck equation:

$$\begin{aligned} \frac{\partial \mathcal{P}(\vec{\alpha})}{\partial t} = & \frac{1}{2\tau} \sum_{\lambda=1}^N \left(\frac{\partial(\alpha_\lambda \mathcal{P}(\vec{\alpha}))}{\partial \alpha_\lambda} + \frac{\partial(\beta_\lambda \mathcal{P}(\vec{\alpha}))}{\partial \beta_\lambda} \right) \\ & + \frac{V}{2} \sum_{\lambda=1}^{N-1} \left\{ -\alpha_{\lambda+1} \beta_{\lambda+1} \left(\frac{\partial(\alpha_\lambda \mathcal{P}(\vec{\alpha}))}{\partial \alpha_\lambda} + \frac{\partial(\beta_\lambda \mathcal{P}(\vec{\alpha}))}{\partial \beta_\lambda} \right) \right. \\ & + (\alpha_\lambda \beta_\lambda + 1) \left(\frac{\partial(\alpha_{\lambda+1} \mathcal{P}(\vec{\alpha}))}{\partial \alpha_{\lambda+1}} + \frac{\partial(\beta_{\lambda+1} \mathcal{P}(\vec{\alpha}))}{\partial \beta_{\lambda+1}} \right) \\ & + 2\alpha_{\lambda+1} \beta_{\lambda+1} \frac{\partial^2 \mathcal{P}(\vec{\alpha})}{\partial \alpha_\lambda \partial \beta_\lambda} \\ & \left. - \frac{\partial^2(\alpha_{\lambda+1} \alpha_\lambda \mathcal{P}(\vec{\alpha}))}{\partial \alpha_{\lambda+1} \partial \alpha_\lambda} - \frac{\partial^2(\beta_{\lambda+1} \beta_\lambda \mathcal{P}(\vec{\alpha}))}{\partial \beta_{\lambda+1} \partial \beta_\lambda} \right\}. \quad (4) \end{aligned}$$

According to the Ito calculus, a Fokker-Planck equation of the form

$$\frac{\partial \mathcal{P}(\vec{\alpha})}{\partial t} = - \sum_n \frac{\partial(f_n(\vec{\alpha}) \mathcal{P}(\vec{\alpha}))}{\partial \vec{\alpha}_n} + \sum_{mm} \frac{\partial^2(M_{mm}(\vec{\alpha}) \mathcal{P}(\vec{\alpha}))}{\partial \vec{\alpha}_n \partial \vec{\alpha}_m} \quad (5)$$

with the symmetric matrix $\mathbf{M}(\vec{\alpha}) = \frac{1}{2} \mathbf{B}(\vec{\alpha}) \mathbf{B}(\vec{\alpha})^T$ is equivalent to the set of Langevin equations [25]:

$$\frac{\partial \vec{\alpha}_n}{\partial t} = f_n(\vec{\alpha}) + B(\vec{\alpha})_{nm} \eta_m, \quad (6)$$

where η_m are independent stochastic Gaussian noise terms, defined by $\langle \eta_m(t) \eta_n(t') \rangle = \delta_{mn} \delta(t - t')$.

Observable quantities are obtained from averaging the Langevin equation over a stochastically generated ensemble. In particular, the average occupations $\langle \hat{a}_\lambda^\dagger \hat{a}_\lambda \rangle = \langle n_\lambda \rangle$ and second order correlations $\langle \hat{a}_\lambda^\dagger \hat{a}_\lambda^\dagger \hat{a}_\lambda \hat{a}_\lambda \rangle$ are given by:

$$\langle \hat{a}_\lambda^\dagger \hat{a}_\lambda \rangle = \langle \beta_\lambda \alpha_\lambda \rangle, \quad (7)$$

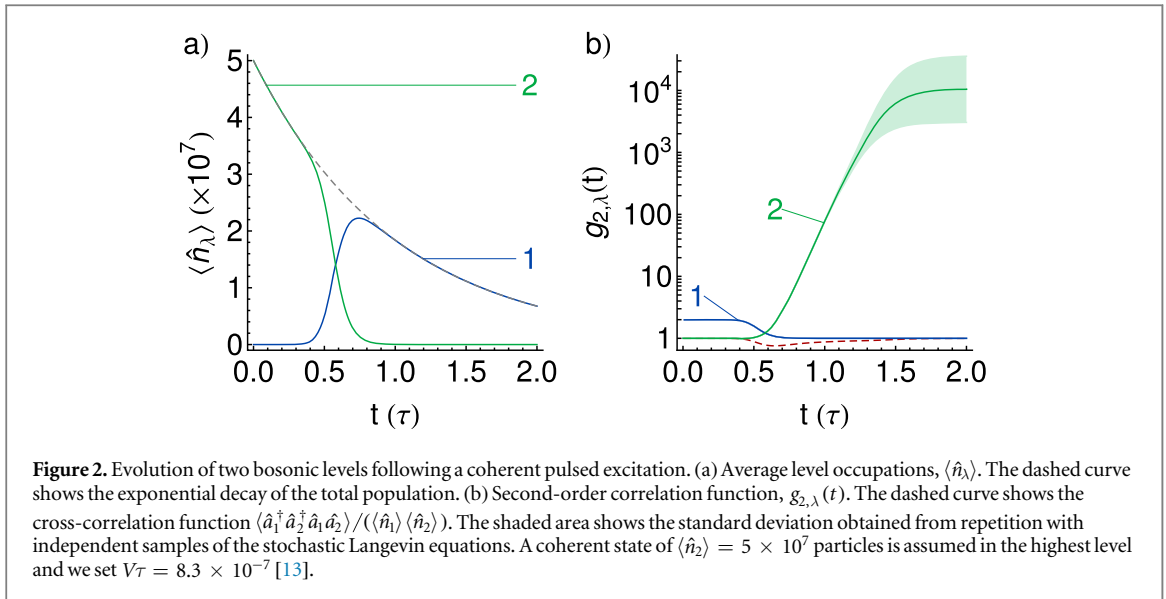
$$\langle \hat{a}_\lambda^\dagger \hat{a}_\lambda^\dagger \hat{a}_\lambda \hat{a}_\lambda \rangle = \langle \beta_\lambda \beta_\lambda \alpha_\lambda \alpha_\lambda \rangle. \quad (8)$$

The normalized second order correlation function is then defined as $g_{2,\lambda\lambda} = \langle \hat{a}_\lambda^\dagger \hat{a}_\lambda^\dagger \hat{a}_\lambda \hat{a}_\lambda \rangle / \langle n_\lambda \rangle^2$. By solving equation (4) numerically, we are able to study the quantum statistical properties of bosonic cascades under conditions of macroscopic occupations, with several millions of particles. The accuracy of the procedure can be assessed by repetition with independent noise samples, from which the standard deviation of different quantities can be deduced.

3. Two-level case

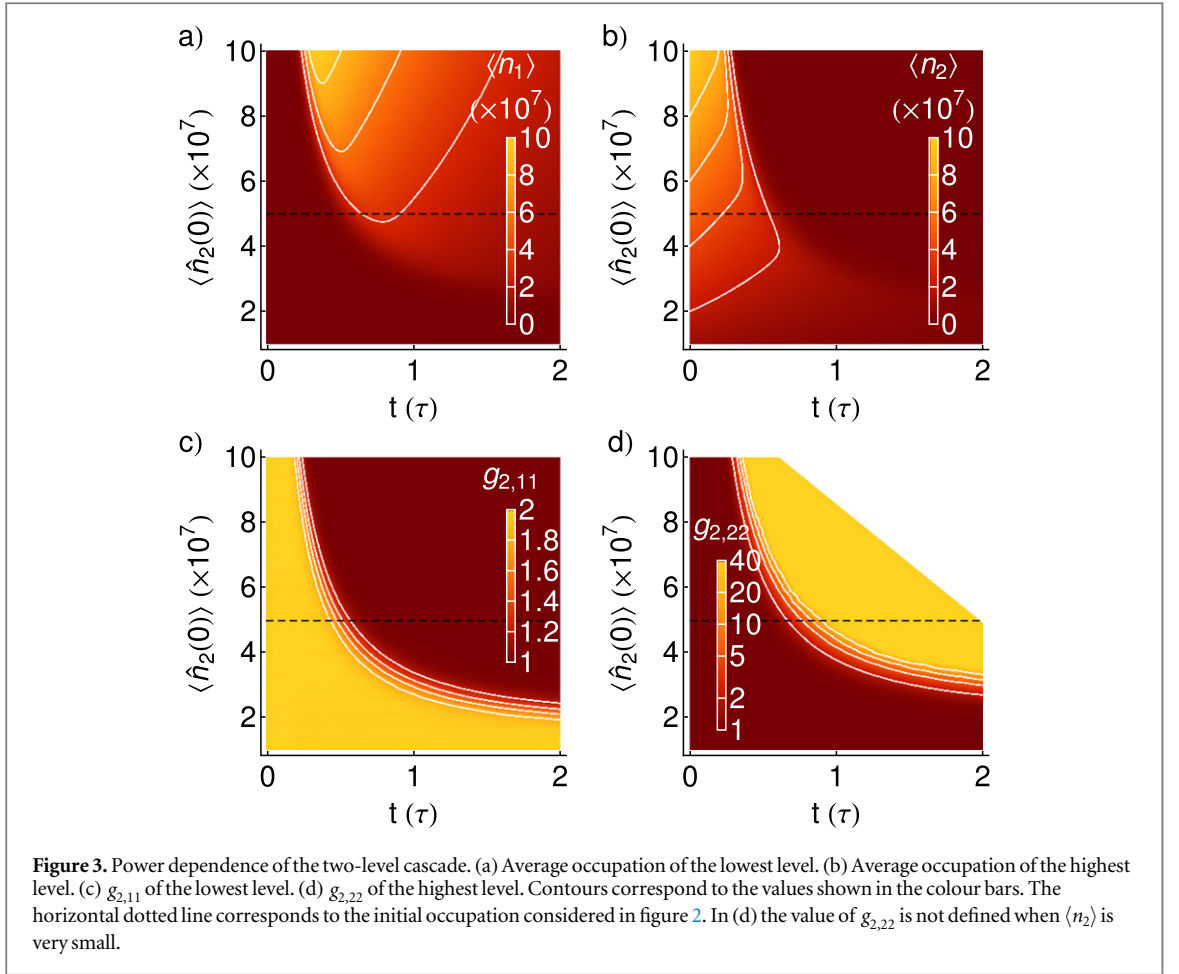
Considering the simplest case of two levels, we obtain the time dependence of the average populations and second order correlation functions shown in figures 2(a) and (b), respectively, for the case where the upper state is initially a coherent state populated with $\langle \hat{n}_2 \rangle = 5 \times 10^7$ particles and the ground state is in the vacuum.

The dynamics of relaxation can be well understood: the population from the upper level decreases in time due to radiative emission and transfers to the lower level. At early times, the upper state populates the ground state in a thermal (or chaotic) state, since it merely provides an incoherent input that increases the average population without developing any other independent observable [26]. Consequently, the particle statistics of the lower level is initially $g_{2,\lambda=1}(t) = 2$, which is the well known second-order correlation for an incoherent gas of bosons. Then, as population increases, stimulated emission sets in and the upper state now empties much faster and predominantly into the ground state. This results in the rapid growth of the ground state population until the upper state is so much depleted that it cannot compensate for the ground state's radiative losses, at



which stage the ground state starts to decay through its own radiative channel. In this buildup phase of the ground state, coherence also grows, as seen in the transition from $g_{2,\lambda=1}(t) = 2$ to $g_{2,\lambda=1}(t) = 1$. Note that the state remains coherent from there on as radiative decay alone does not dephase the condensate. More striking is the statistics of the upper state. While its second-order correlation function was initially unity, as befits a coherent state, and remained essentially unaffected in the first phase of radiative decay and spontaneous emission into the ground state, there is a pronounced superbunching $g_{2,\lambda=1}(t) > 2$ that forms when the state is rapidly emptied. Remarkably, just as the statistics of the ground state remains equal to one until complete evaporation, the upper state's statistics also remains pinned at the value it reached when it completed its transfer. This is thanks to the absence of back scattering from the lower to the upper mode, negligible in absence of a THz cavity and at the low temperatures we consider. In such a case, detailed-balance breaks down and while the steady state remains unique (and also trivial, namely, the vacuum), it is approached in different ways depending on the initial condition. We will consider here mainly coherent states as they are experimentally the most relevant ones, and are theoretically sufficient to evidence the effect. The well defined and high value of $g_{2,\lambda}(t)$ for a state asymptotically approaching the vacuum is due to the mathematical limit of the two vanishing quantities (see equations (7) and (8)) that exists even for arbitrarily small probabilities of occupation. In practice, numerical simulations are less stable in this region and experimental measurements would also be increasingly difficult. Nevertheless, the most interesting phenomenology is the superbunching that develops in the phase where the upper state quickly empties into the ground state, at a time when there are still a macroscopic number of particles. In this process, where one condensate is sucked by another one, the photons emitted radiatively by the upper state will indeed exhibit a bright superbunched statistics.

The reason for this peculiar superbunching behaviour is to be found in the mechanism of coherence buildup. Equation (1) is equivalent to a quantum Boltzmann master equation [27], in which formalism coherence—as measured by Glauber's correlation functions—builds up thanks to population correlations developed by the dynamics, even in the absence of interactions or external potentials. Note that Boltzmann master equations alone (without retaining particle number correlations) do not yield such effects. In the conventional case of Bose condensation, the Poisson fluctuations of a single condensed state (usually the ground state), which leads to $g_n = 1$ for all n , are due to this single state acquiring the fluctuations of a macroscopic system [28]. Since the central limit theorem states that the distribution of a large number of random variables (the excited states) is a Normal distribution—very close to the Poisson fluctuations of a coherent state for large populations—so does also fluctuate the condensate in one single state (the ground state). Each excited state taken in isolation contributes very little to the condensate in a macroscopic system and its statistical properties in isolation of the other excited states are not significantly altered. In bosonic cascades, however, the asymmetry between ground and excited states is lost since there can be a few macroscopically occupied excited states. This is a peculiar configuration where coherence is acquired from another single coherent state, rather than from a macroscopic ensemble of weakly occupied thermal states (the statistical properties of which do not matter, still following the central limit theorem, as long as they obey generic conditions of independence and convergence). This peculiarity is the reason why a single excited state can develop such a pronounced superbunching when acting as a reservoir for another condensate to grow in another single state. The fast loss of population from a



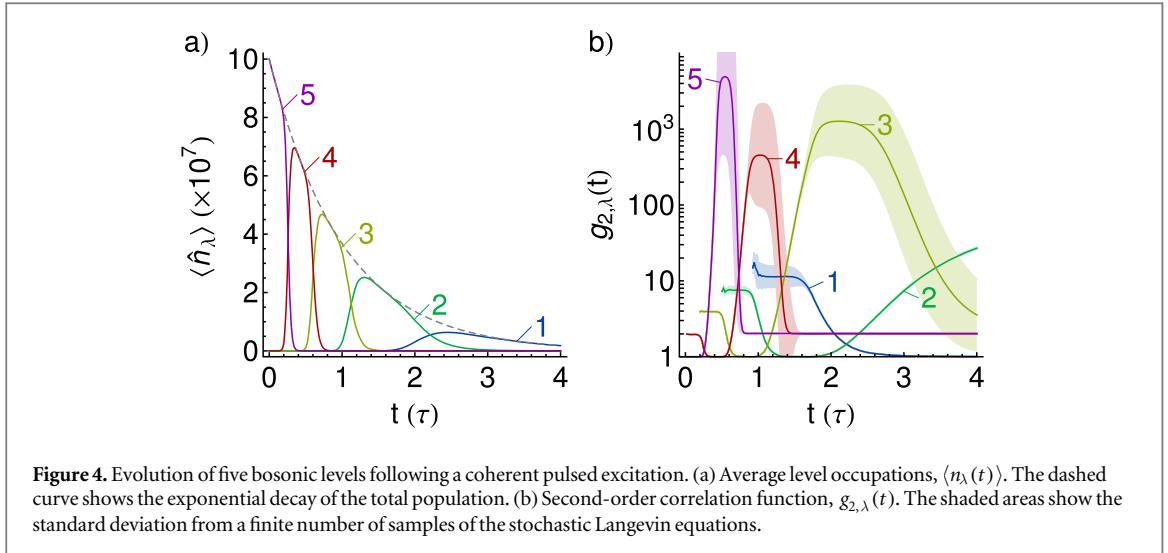
single state to provide for the coherence of another single state, results in the superbunching, or extreme chaos, for the provider that substitutes a macroscopic environment. This interpretation is supported by the following facts: the superbunching increases with both larger initial populations and $V\tau$, i.e., is favoured by long-lived particles and fast relaxation rates of macroscopic states, that imprint larger correlations when the condensate transfers between the two modes. The other important specificity is the absence of backscattering, which would tame the superbunching of the excited state and eventually bring its statistics to that of a thermal state. In its absence, the superbunching reaches a maximum value that stays pinned forever, giving rise to the plateau, although as time passes, less particles are available to manifest this. The dependence of the second order correlation functions, together with the occupation numbers, on the initial occupation number is illustrated in figure 3. The lowest mode maintains the behaviour of being initially thermal, but then becomes a coherent state upon being highly populated, as the upper state makes the transition from coherent to super-bunched light. Note that the effect is robust to dephasing (see appendix B), as long as it does not destroy correlations between the states and thus prevent the transfer of coherence of the condensate, to transfer its population only. In the next section, we also show how the effect weakens with additional levels. In an ideal system, the superbunching is a very strong effect. The greatest superbunching is obtained with non-fluctuating quantum states. For instance, with a Fock state of two particles as the initial condition, i.e., $p(n, m, 0) = \delta_{n,0}\delta_{m,2}$, one finds (see appendix A):

$$g_{2,2}(t) = \frac{(1 + V\tau)^4 e^{2(1+V\tau)t/\tau}}{2(V^2\tau(V\tau t + t + \tau) + (1 + 2V\tau)e^{(1+V\tau)t/\tau})^2}, \quad (9)$$

the limit of which as $t \rightarrow \infty$ provides the superbunching plateau in this case:

$$g_{2,2}(t) = \frac{(1 + V\tau)^4}{2(1 + 2V\tau)^2}. \quad (10)$$

Equation (10) goes from $1/2$ (the $g_{2,2}(t)$ of the initial condition) when $V\tau \rightarrow 0$ up to ∞ when $V\tau \rightarrow \infty$. For the value of $V\tau$ we considered in figure 2, no dramatic effect results with two particles, and the large population of a condensate is required instead to bring in superbunching (as well as to pass the condensation threshold). There are nevertheless several remarkable features of this Fock state solution. Even though the system converges towards the vacuum, this closed-form expression proves that the photon statistics converges to a well-defined,



finite value: the superbunching plateau. Moreover, this plateau is unbounded with $V\tau$, so an arbitrarily high bunching can be obtained with two particles only, provided their lifetime and scattering rate are large enough. In particular, equation (10) shows that particles with infinite lifetimes exhibit a divergent bunching. This remains true for more particles in a Fock state but not, interestingly, for the correlations developed by a coherent state. These are not strong enough to result in such a divergence, and a plateau is obtained also for infinitely lived particles. The plateau is still extremely large, namely, for particles with infinite lifetime, it grows like the exponential of the population, $g_2 = \exp(n_2)$, for an initial coherent state of n_2 particles (the proof is given in appendix A). Taking the case of figure 2 of the main text, we deal with $V\tau = 8.3 \times 10^{-7}$ and $n_2 = 8.3 \times 10^7$, thus reducing to a mere few hundreds the mind boggling superbunching $g_{2,2} \approx 10^{21714724}$ that would result for infinitely lived particles. While the superbunching is drastically reduced by lifetime, it still allows to produce a significant superbunching involving millions of particles with today's experimentally accessible values of $V\tau \ll 1$.

This mechanism of superbunching is different to the formation of superbunching in chaotic lasers [19]. While there may also be a transfer of coherence between competing modes, those systems require the engineering of additional feedback. Random lasers also offer an alternative route to superbunching, but here we achieve very high levels of superbunching without the necessity of a nonlinear chaotic system in the first place. We note that we are unaware of any classical model that can describe the aforementioned effect, while we do not rule out such a possibility.

4. Multi-level case

When there are more states in the cascade, the superbunching is weaker due to the greater resemblance of the bosonic cascade from a single condensate and a macroscopic reservoir: the more levels there are and/or the less they are occupied, the more the system resembles the usual scenario. However, the statistics then exhibits a richer dynamics as the condensate undergoes several stages of transfers. Considering a larger cascade composed of five levels, still with a macroscopic occupation of the highest excited state ($\langle \hat{n}_{\lambda=5} \rangle(t=0) = 10^8$) and all others empty, we obtain the result shown in figure 4. Looking first at the populations (figure 4(a)), we see that there is a staggered oscillation in the successive levels, corresponding to the transfers of particles down through each level of the cascade. The beginning of the relaxation, between the two-highest excited states, is similar to the case already studied, but since the recipient for the highest excited state is also the source for another state below, the dynamics is echoed down the ladder until it reaches the final ground state. In the process, the dynamics slows down and is tamed in intensity, as particles are constantly lost to the environment. The second order correlation functions also exhibit a staggering effect, which can be understood as a generalization of the behaviour observed in figure 2 for the two-level case, but with a richer phenomenology.

We now describe the more complex superbunching observed in the multi-level case. The feeding condensate is sucked into the growing one but this process gets disturbed when the latter condensate becomes in its turn the feeding singly occupied macroscopic state that trades its coherence to the state below it. This releases the drain on the former condensate and relaxes the superbunching of its remaining particles. In the phase of condensation, the state that grows its coherence does so not only at the expense of its exciting state, but also of the state below. This results in a smaller plateau of superbunching for the latter state that sandwiches the

condensate together with the plateau of large superbunching from the exciting state. As a result, the particle statistics for each state is an intricate sequence of several plateaus joined by abrupt jumps as the condensate starts to form or starts to empty. For a generic state—that is, one that is not too close from the most excited state or from the ground state—the statistical relief has five plateaus: (i) a thermal state starting with, and growing from, the vacuum before the cascade is started; (ii) a plateau of ‘small super-bunching’ as it provides coherence from below to its exciting state; (iii) a coherent plateau as the state is building its own coherence from the excited state now in its process of avalanche; (iv) a plateau of ‘big superbunching’ as the state feeds the state below, becoming super-chaotic in the transfer of coherence; and finally, (v) a thermal state as the process got transported to states below, with the next state now in its phase of ‘big superbunching’ and that two states below in that of building its coherence. The most relevant plateau is that of stage (iv), as it is present for all the excited states, has the stronger superbunching and the stronger signal. All the excited states except that immediately above the ground state ultimately revert to a thermal state, once the peculiar dynamics of bosonic cascade is long gone. The complex concordance of these several stages in statistics with the corresponding ones in populations can be observed in figure 4(b), showing the beautiful and peculiar dynamics of particle correlations in bosonic cascades.

5. Conclusions

We reported a mechanism based on a ladder of bosonic condensates coupled as nearest-neighbours to produce extremely high values of photon bunching, scaling in an ideal limit as the exponential of the initial population. With stochastic Langevin equations, we can compute the quantum statistics of such bosonic cascades with several levels and for macroscopic occupations, showing how a bright superbunched signal can still be obtained for realistic experimental parameters. The peculiar nature of this system, where coherence is transferred between single states, accounts for the superbunching, which develops in the phase where one condensate empties suddenly into the state below. The observation of this strong and striking effect could be made in wide parabolic quantum wells where excitons are confined as whole particles. In addition to illustrating the fundamental features of coherence buildup in non-interacting bosonic gases, this mechanism could act as a resource for ghost imaging applications based on bright superbunched optical fields.

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Appendix A. Closed-form solutions

In this appendix we derive some exact results for particular cases, to support our interpretation and prove that the remarkable dynamics we find numerically is not an artifact of the calculation, but can be tracked down to analytical expressions. We first consider the effect reduced at its simplest expression: a Fock state of two particles only as the initial state. The master equation then reads simply

$$\partial_t \hat{\rho} = V(2\hat{b}\hat{a}^\dagger \hat{\rho} \hat{b}^\dagger \hat{a} - \hat{b}^\dagger \hat{b} \hat{a} \hat{a}^\dagger \hat{\rho} - \hat{\rho} \hat{b}^\dagger \hat{b} \hat{a} \hat{a}^\dagger) + (\mathcal{L}_a + \mathcal{L}_b) \hat{\rho} / \tau \text{ with } \mathcal{L}_c = 2\hat{c}\hat{\rho}\hat{c}^\dagger - \hat{c}^\dagger\hat{c}\hat{\rho} - \hat{\rho}\hat{c}^\dagger\hat{c}.$$

In contrast to the cases dealt with in the main text that involve a very large number of particles, calling for dedicated techniques, with few particles it is possible, and simpler, to solve directly for the diagonal elements of the density matrix $p_{nm} = \langle n, m | \hat{\rho} | n, m \rangle$, which yields the joint probability of having n particles in the ground state and m in the excited state. Importantly, here, p_{nm} does not factorize as a product of probabilities, in which case no coherence would grow in the system and only a transfer of population as described by Boltzmann equations would result. The correlated equation of motion is easily obtained from the master equation as

$$\begin{aligned} \partial_t p_{nm}(t) = & -p_{nm}(t)[V(n+1)m + (n+m)/\tau] + p(n-1, m+1, t)V(m+1)n \\ & + p(n+1, m, t)(n+1)/\tau + p(n, m+1, t)(m+1)/\tau \end{aligned}$$

in the small configuration space $0 \leq n+m \leq 2$, which allows a closed-form solution for all six probabilities $p_{nm}(t)$, e.g., with two particles in the excited state at $t=0$, $p(0, 0, t) = \exp(-2t/\tau)(\exp(t/\tau) - 1)^2$ (note that the probability for the vacuum is V -independent) or $p(0, 2, t) = \exp(-2t(1+V\tau)/\tau)$, etc., from which one can obtain the result in equation (9). Fock states with more particles lead to similar results but with expressions too awkward to be written here and bringing little additional insights.

We can also address some cases with an arbitrary initial state in the limit of infinite lifetime (hence only relaxation). The master equation reads simply $\partial_t \rho = V(2\hat{b}\hat{a}^\dagger\rho\hat{b}^\dagger\hat{a} - \hat{b}^\dagger\hat{b}\hat{a}\hat{a}^\dagger\rho - \rho\hat{b}^\dagger\hat{b}\hat{a}\hat{a}^\dagger)$ and, in the Fock state basis, $\partial_t p_{nm}(t) = -p_{nm}(t)V(n+1)m + p(n-1, m+1, t)Vn(m+1)$. The key here is that the equation is closed in the manifold $n+m = \mu$ of constant excitations. We can then write the equation for the vector \mathbf{v} with the elements p_{nm} arranged in groups $n+m = \mu = \text{cste}$:

$$\mathbf{v} = \begin{pmatrix} p_{00} \\ p_{01} \\ p_{10} \\ p_{02} \\ p_{11} \\ p_{20} \\ \vdots \\ p_{0,\mu} \\ p_{1,\mu-1} \\ p_{2,\mu-2} \\ \vdots \\ p_{\mu-1,1} \\ p_{\mu,0} \\ \vdots \end{pmatrix} \quad (A.1)$$

as $\partial_t \mathbf{v} = MVt\mathbf{v}$ where M is in block-diagonal form:

$$M = \left(\begin{array}{cccc|cccc|cccc|cccc} \hline 0 & & & & & & & & & & & & & & & & & & \\ \hline & -1 & 0 & & & & & & & & & & & & & & & & \\ \hline & & 1 & 0 & & & & & & & & & & & & & & & \\ \hline & & & & -2 & 0 & 0 & & & & & & & & & & & & \\ \hline & & & & & 2 & -2 & 0 & & & & & & & & & & & \\ \hline & & & & & & 0 & 2 & 0 & & & & & & & & & & \\ \hline & & & & & & & & & \ddots & & & & & & & & & \\ \hline & & & & & & & & & & -\mu & 0 & 0 & \dots & 0 & 0 & & & \\ \hline & & & & & & & & & & \mu & -2(\mu-1) & 0 & \dots & 0 & 0 & & & \\ \hline & & & & & & & & & & 0 & 2(\mu-1) & -3(\mu-2) & \dots & 0 & 0 & & & \\ \hline & & & & & & & & & & \vdots & \dots & \vdots & \ddots & \vdots & \vdots & & & \\ \hline & & & & & & & & & & 0 & 0 & 0 & 2(\mu-1) & -\mu & 0 & & & \\ \hline & & & & & & & & & & 0 & 0 & 0 & 0 & \mu & 0 & & & \\ \hline & & & & & & & & & & \vdots & & & & & & \ddots & & \\ \hline \end{array} \right) \quad (A.2)$$

The solution of $\partial_t \mathbf{v} = Vt \text{diag}(M_0, M_1, \dots, M_\mu, \dots)\mathbf{v}$ being simply:

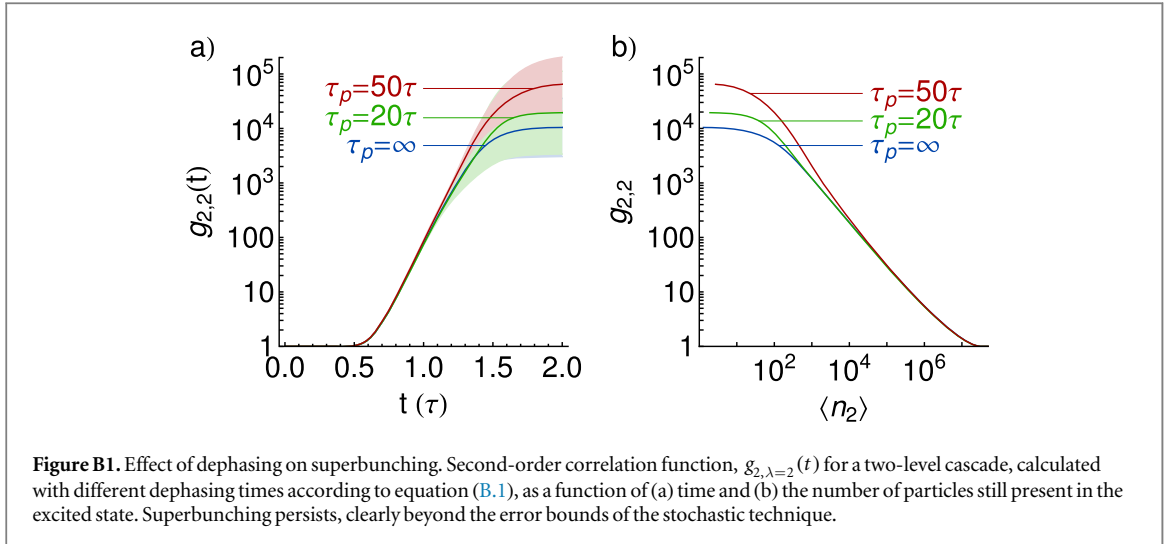
$$\mathbf{v}(t) = \text{diag}(\exp(V\mathbf{M}_0t), \exp(V\mathbf{M}_1t), \dots, \exp(V\mathbf{M}_\mu t), \dots)\mathbf{v}(0), \quad (A.3)$$

with, beside, $\mathbf{v}(0)$ only having the first elements $p_{0m} \equiv p(m)$ of each block nonzero (since we assume that only the excited state is initially populated), the problem is reduced to calculating the first column of the matrix exponential of a sparse matrix of the form spelled out in equation (A.2). The reduced probability for the excited state alone $p_m(t)$ is then obtained by summing all the elements of the form p_{nm} for fixed m and varying n , of which there is one in each block starting from the m th:

$$p_m(t) = \sum_{\mu=m}^{\infty} [\exp(V\mathbf{M}_\mu t)]_{1,1+\mu-m} p(m). \quad (A.4)$$

This is still an awkward problem for arbitrary times with a voluminous general solution (that is, however, straightforward to compute numerically). But since we are interested in the long term behaviour, we can simplify the solution by retaining only the dominant terms. Note that it is not enough to aim for a steady state solution, that is trivially the vacuum, which we have already commented is approached differently depending on the initial state.

We therefore proceed with the calculation of the leading term of $\exp(V\mathbf{M}_\mu t)$. Since \mathbf{M}_μ is triangular, its eigenvalues appear directly on the diagonal, i.e., they are $V(n+1)(\mu-n)t$. Note that one of them is zero. From the Jordan decomposition, one obtains that the terms of $\exp(V\mathbf{M}_\mu t)$ are linear combinations of $(Vt)^k \exp(Vt(n+1)(\mu-n))$ for integer k (possibly zero). We can exclude the non-decaying terms for p_{m0} on physical grounds, since the probability must tend to zero with time for the excited state. As a conclusion, the leading term for the μ th block is that given by the smallest nonzero eigenvalue, which is that of order μ , i.e., also the term that weights $p_{0\mu}$. Provided that no probability be exactly zero (which excludes Fock states or similar



from the following, but includes Gaussian states including thermal and coherent), one can therefore simplify equation (A.4), for sufficiently large t , as:

$$p_m(t) = \exp(-mt)p(m), \quad (\text{A.5})$$

since all the terms in the sum after the first one are at least of a higher exponential order (possibly with also t^k factors that do not, however, lead over the exponential of smaller decay rate). The only term that is not straightforward to obtain in this way is the vacuum, since it has contributions from the non-decaying eigenvalues that cannot be easily simplified. It is, however, directly obtained by normalization, $p_0(t) = 1 - \sum_{m=1}^{\infty} \exp(-mt)p(m)$. As a conclusion of the above, if the initial state is a coherent state, with $p(m) = \exp(-\alpha^2/2)\alpha^{2m}/m!$, one finds $g_2 = \exp(\alpha^2(1 - e^{-t}))$ at large enough times, which tends to $\exp(\alpha^2)$, as stated in the text. A thermal state as initial condition would provide a superbunching that grows only linearly with the number of particles: $g_2 = 2(n + 1)$. Note that since Fock states cancel the leading terms, their long t behaviour cannot be inferred in this way. It can be computed, instead, from the full solution, e.g., equation (9) shows that for infinitely lived particles, the long-term behaviour of the g_2 for a Fock state of two particles is $\exp(2t)/(2t^2)$ (and indeed diverges instead of producing a plateau).

Appendix B. Effect of dephasing

The effect of dephasing on the predicted superbunching can be modeled by an additional term added to the right-hand side of equation (1):

$$\frac{1}{2\tau_p} \sum_{\lambda=1}^N ([\hat{a}_{\lambda}^{\dagger} \hat{a}_{\lambda}, \hat{\rho} \hat{a}_{\lambda}^{\dagger} \hat{a}_{\lambda}] + [\hat{a}_{\lambda}^{\dagger} \hat{a}_{\lambda} \hat{\rho}, \hat{a}_{\lambda}^{\dagger} \hat{a}_{\lambda}]), \quad (\text{B.1})$$

where τ_p characterizes the dephasing time.

The dynamics in the presence of dephasing can again be calculated with the stochastic treatment outlined in section 2, with appropriate modifications. Figure B1 shows the effect of dephasing on a two-level cascade, where we find that significant superbunching persists within the accuracy of our method.

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