Strongly interacting photons in arrays of dissipative nonlinear cavities under a frequency-dependent incoherent pumping

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We report a theoretical study of a quantum optical model consisting of an array of strongly nonlinear cavities incoherently pumped by an ensemble of population-inverted two-level atoms. Projective methods are used to eliminate the atomic dynamics and write a generalized master equation for the photonic degrees of freedom only, where the frequency-dependence of gain introduces non-Markovian features. In the simplest single cavity configuration, this pumping scheme allows for the selective generation of Fock states with a well-defined photon number. For many cavities in a weakly non-Markovian limit, the non-equilibrium steady state recovers a Grand-Canonical statistical ensemble at a temperature determined by the effective atomic linewidth. For a two-cavity system in the strongly nonlinear regime, signatures of a Mott state with one photon per cavity are found.

I. INTRODUCTION

The study of quantum many-body systems is one of the most active fields of modern condensed-matter physics. Among the most celebrated effects, we can mention frictionless flows in superfluid and superconducting systems and the geometrical quantization features of the fractional quantum Hall effect. While this physics was traditionally studied in liquid Helium samples \([1,2]\), in atomic nuclei \([3]\), in quark-gluon plasmas \([29,30]\), or in electron gases confined in solid-state devices \([3,4,9,10]\), the last two decades have witnessed impressive advances using ultra-cold atomic gases trapped in magnetic or optical traps \([6,8]\).

In the last few years, a growing community has started investigating many-body effects in the novel context of the so-called quantum fluids of light \([17]\), i.e. assemblies of many photons confined in suitable optical devices, where effective photon-photon interactions arise from the optical nonlinearity of the medium. After the pioneering studies of Bose-Einstein condensation \([18]\) and superfluidity \([19]\) effects in dilute photon gases in weakly nonlinear media, a great interest is presently being devoted to strongly nonlinear systems, where even single photons are able to appreciably affect the optical properties of the system.

The most celebrated example of such physics is the photon blockade effect \([16]\), where the presence of a single photon in a cavity is able to detune the cavity frequency away from the pump laser, so that photons behave as effectively impenetrable particles. Experimental realizations of this idea have been reported by several groups using very different material platforms, from single atoms in macroscopic cavities \([12]\), to single quantum dots in photonic crystal cavities \([12,13]\), to single Josephson qubits in circuit QED devices for microwaves \([14]\).

Scaling up to arrays of many cavities coupled by photon tunneling is presently a hot challenge in experimental physics, as it would realize a Bose-Hubbard model for photons where the photon blockade effect may lead to a rich physics, including the superfluid to Mott-insulator phase transition at a commensurate filling or Tonks-Girardeau gases of impenetrable photons in one-dimensional continuum models. The first works on strongly correlated photons were restricted to quasi-equilibrium regimes where the photon loss rate is much slower than the internal dynamics of the gas so that the system has time to thermalize and/or be adiabatically transferred to the desired strongly correlated state \([21,22]\). While this assumption might be satisfied in suitably designed circuit-QED devices in the microwave domain, radiative losses are hardly negligible in realistic optical cavities in the infrared or visible domain, so that thermalization is generally far from being granted \([11,17]\).

As a result, a very active attention has been recently devoted to the peculiar non-equilibrium effects that arise for realistic loss rates. Starting from the pioneering work on photon blockade in non-equilibrium photonic Josephson junctions \([28]\), the interest has been focused on the study of schemes to generate strongly correlated many-body states in the very non-equilibrium context of photon systems, where the steady-state is not determined by a thermal equilibrium condition, but by a dynamical balance of driving and losses.

The first such scheme proposed in \([23]\) was based on a coherent pumping: provided the different many-body states are sufficiently separated in energy, many-photon processes driven by the coherent external laser are able to selectively address each many-body state as done in optical spectroscopy of atomic levels. In this way, the non-equilibrium condition is no longer just a hindrance, but offers new perspectives, as it allows to individually probe each excited state. Furthermore,
the appreciable radiative losses make microscopic information on the many-body wavefunction be directly encoded in the quantum coherence of the secondary emission from the device [24, 31, 32]. While this coherent pumping scheme offers a viable way to generate and control few photon states in small arrays, its efficiency is restricted to mesoscopic systems where the different states are well-separated in energy. Moreover, this scheme intrinsically leads to coherent superpositions of states of different photon number: while this feature is intriguing in view of observing many-body braiding phases [33], it is not ideally suited to generate states with a well-defined photon number such as Mott-insulator states.

The identification of new schemes that do not suffer from these limitations is therefore of great importance in view of experiments. In the present work we study the potential of frequency-dependent gain processes to selectively generate strongly correlated states of photons in arrays of strongly non-linear cavities. The frequency-dependence of amplification is a well-known fact of laser physics and is often exploited to linear cavities. The frequency-dependence of amplification is strongly correlated states of photons in arrays of strongly non-frequency-dependent gain processes to selectively generate experiments. In the present work we study the potential of positions of states of different photon number: while this feature is intriguing in view of observing many-body braiding phases [33], it is not ideally suited to generate states with a well-defined photon number such as Mott-insulator states.

In the last years, similar questions have been theoretically addressed by several groups. Just to mention a few of them, a scheme to obtain a thermal state at finite temperature with a non-vanishing effective chemical potential for photons has been proposed in [24] using a clever parametric system-bath coupling. In particular, the potential of this configuration in view of generating strongly correlated photon states in circuit-QED and opto-mechanical context was assessed. With respect to this proposal and to the engineered dissipations originally proposed in [33] in the atomic context and then extended to the photonic context in [34], our approach is based on a quite commonly observed feature of photonic systems such as a frequency-dependent gain: on one hand, engineering a frequency-dependent gain may open new avenues to optimize the performance of optical devices; on the other hand, the role of frequency-dependent gain in photon and polariton Bose-Einstein condensation experiments [18, 35, 36] is presently under active study [23, 37]. Finally, a pioneering discussion of collective coherence in arrays of cavities embedding single population-inverted two-level atoms has recently appeared in [39], but little attention was paid to the effect of strong non-linearities.

The structure of the article is the following. In Sec III we present the physical system and we develop the theoretical model based on a master equation for the cavities coupled to the atoms of the gain medium. The projective method to eliminate the atomic degrees of freedom and write a master equation for the photonic density matrix is sketched in Sec III. First application of the method to a single cavity configuration is discussed in Sec IV and specific features of the weak and the strong nonlinearity cases are illustrated, e.g. a novel mechanism for optical bistability and the selective generation of Fock states with a well defined photon number. The richer physics of many cavity arrays is discussed in Sec V in a weakly non-Markovian regime, an effective Grand-Canonical distribution is obtained even in the absence of thermalization mechanisms; in a strongly nonlinear and non-Markovian regime, signatures of a Mott insulator state with one photon per cavity are illustrated. Conclusions are finally drawn in Sec VI. In the Appendices, we provide the details of the derivation of the photonic master equation using projective methods, on the exact stationary state in the Markovian case, and on a perturbative expansion of the coherences in the weakly non-Markovian limit.

II. THE PHYSICAL SYSTEM AND THE THEORETICAL MODEL

In this work, we consider a driven-dissipative Bose-Hubbard model for photons in an array of \( k \) coupled nonlinear cavities [17]. The Hamiltonian for the isolated system dynamics has the usual form

\[
H_{\text{ph}} = \sum_{i=1}^{k} \left[ \Omega_{\text{cav}} a_i^\dagger a_i + \frac{U}{2} a_i^\dagger a_i^\dagger a_i a_i - \sum_{(i,j)} \hbar J a_i^\dagger a_j^\dagger + \hbar c \right] : \tag{2.1}
\]

all \( k \) cavities have the same natural frequency \( \Omega_{\text{cav}} \). They are arranged in a one-dimensional geometry and are coupled via tunneling processes with amplitude \( J \). Each cavity is assumed to contain a \( \chi^{(3)} \) Kerr nonlinear medium, which induces effective repulsive interactions between photons in the same cavity.

Dissipative phenomena due the finite transparency of the mirrors and absorption by the cavity material are responsible for a finite lifetime of photons, which naturally decay at a rate \( \Gamma_{\text{loss}} \). As mentioned in the introduction, the key novelty of this work consists in the mechanism that is proposed to compensate for losses and replenish the photon population. Instead of a coherent pumping or a very broad-band amplifying laser medium, we consider a configuration where a set of \( N_{\text{at}} \) two level atoms is present in each cavity. Each atom is strongly pumped at a rate \( \Gamma_{\text{pump}} \), spontaneously decays to its ground state at a rate \( \gamma \) and, most importantly, is coupled to the cavity with a Rabi frequency \( \Omega_R \). The free evolution of the atoms and their coupling to the cavities are described by the following Hamiltonians,

\[
H_{\text{at}} = \sum_{i=1}^{k} \sum_{l=1}^{N_{\text{at}}} \omega_{\text{at}} a_i^\dagger a_i \sigma_i^{-(l)} : \tag{2.2}
\]

\[
H_{\text{It}} = \Omega_R \sum_{i=1}^{k} \sum_{l=1}^{N_{\text{at}}} \left[ a_i^\dagger \sigma_i^{-(l)} + a_i \sigma_i^{+(l)} \right] : \tag{2.3}
\]
the atomic frequency $\omega_{at}$ is assumed to be in the vicinity (but not necessarily resonant) with the cavity mode. The atom-cavity coupling is assumed to be weak enough $\Omega_R \ll \omega_{at}, \omega_{cav}$ to be far from the ultra-strong coupling regime.

As usual, the dissipative dynamics under the effect of the pumping and decay processes can be described in terms of a master equation for the density matrix $\rho$ of the whole atom-cavity system,

$$\partial_t \rho = \frac{1}{i} [H_{ph} + H_{at} + H_I, \rho] + \mathcal{L}(\rho), \quad (2.4)$$

where the different dissipative processes are summarized in the Lindblad super-operator $\mathcal{L} = \mathcal{L}_{pump} + \mathcal{L}_{loss, at} + \mathcal{L}_{loss, cav}$, with

$$\mathcal{L}_{pump} = \frac{\Gamma_{pump}}{2} \sum_{i=1}^{k} \sum_{l=1}^{N_{at}} \left[ 2\sigma_i^{+(l)} \rho \sigma_i^{-(l)} + \sigma_i^{-(l)} \sigma_i^{+(l)} \rho - \rho \sigma_i^{+(l)} \sigma_i^{-(l)} \right], \quad (2.5)$$

$$\mathcal{L}_{loss, at} = \frac{\gamma}{2} \sum_{i=1}^{k} \sum_{l=1}^{N_{at}} \left[ 2\sigma_i^{-(l)} \rho \sigma_i^{+(l)} + \sigma_i^{+(l)} \sigma_i^{-(l)} \rho - \rho \sigma_i^{+(l)} \sigma_i^{-(l)} \right], \quad (2.6)$$

$$\mathcal{L}_{loss, cav} = \frac{\Gamma_{loss}}{2} \sum_{i=1}^{k} \left[ 2a_i \rho a_i^\dagger - a_i^\dagger a_i \rho - \rho a_i^\dagger a_i \right]. \quad (2.7)$$

III. CLOSED MASTER EQUATION FOR THE PHOTONIC DENSITY MATRIX

If the atom-cavity coupling $\Omega_R$ is small enough compared to the atomic pumping rate, the atomic population is concentrated in the excited state and it is possible to use projective methods to write a closed master equation for the photonic density matrix where the atomic degrees of freedom $B$ have been traced out, $\rho_{ph} = Tr_B \rho$. As calculations are a bit cumbersome, the interested reader can find the full derivation in Appendix A. The resulting photonic master equation then reads

$$\partial_t \rho_{ph} = -i [H_{ph}, \rho_{ph}(t)] + \mathcal{L}_{loss, cav} + \mathcal{L}_{em, cav}, \quad (3.1)$$

with

$$\mathcal{L}_{loss} = \frac{\Gamma_{loss}}{2} \sum_{i=1}^{k} \left[ 2a_i \rho a_i^\dagger - a_i^\dagger a_i \rho - \rho a_i^\dagger a_i \right], \quad (3.2)$$

$$\mathcal{L}_{em} = \frac{\Gamma_{em}}{2} \sum_{i=1}^{k} \left[ a_i^\dagger \rho a_i + a_i^\dagger \rho a_i - a_i^\dagger a_i \rho - \rho a_i^\dagger a_i \right], \quad (3.3)$$

describing photonic losses and emission processes, respectively. While the loss term has a standard Lindblad form at rate $\Gamma_{loss}$, the emission term keeps some memory of the atomic dynamics as it involves modified lowering and raising operators

$$\tilde{a}_i = \frac{\Gamma_{pump}}{2} \int_0^\infty d\tau e^{-i\omega_{at}\tau - \Gamma_{pump}/2} a_i(-\tau), \quad (3.4)$$

$$\tilde{a}_i^\dagger = [\tilde{a}_i]^\dagger \quad (3.5)$$

which contain the photonic (hamiltonian and dissipative) dynamics during pumping. In the limit we are considering in which photonic losses are slow with respect to atomic pump, these operators are the interaction picture ones with respect to the photonic hamiltonian in the cavity array and have a simpler expression:

$$a_i(\tau) = e^{iH_{ph}\tau} a_i e^{-iH_{ph}\tau}. \quad (3.6)$$

The Fourier-like integral in Eqs (3.4) and (3.5) is responsible for the frequency selectivity of the emission, as the integral is maximum when the free evolution of $a_i$ occurs at a frequency close to the atomic one $\omega_{at}$. The peak emission rate when exactly on resonance is equal to

$$\Gamma_{em} = \frac{4N_{at}^2 \Omega_R^2}{\Gamma_{pump}}; \quad (3.7)$$

under the small $\Omega_R$ assumptions of our theory, for a finite atom number $N_{at}$, one is bound to have $\Gamma_{em} \ll \Gamma_{pump}$.

A deeper physical insight on these operators can be obtained by looking at their matrix elements in the basis of eigenstates of the photonic hamiltonian. We consider two eigenstates $|f\rangle$ (resp. $|f'\rangle$) with $N$ (resp. $N + 1$) photons and energy $\omega_f$ (resp. $\omega_{f'}$). After elementary manipulation,
we see that the emission amplitude follows a Lorentzian law
\[
\langle f | \tilde{a}_i | f' \rangle = \frac{\Gamma_{pump}/2}{i(\omega_{at} - \omega_{f,f}) + \Gamma_{pump}/2} \langle f | a_i | f' \rangle \tag{3.8}
\]
\[
\langle f' | \tilde{a}_i^\dagger | f \rangle = \frac{\Gamma_{pump}/2}{-i(\omega_{at} - \omega_{f,f}) + \Gamma_{pump}/2} \langle f' | a_i^\dagger | f \rangle \tag{3.9}
\]
as a function of the detuning $\omega_{f,f} = \omega_f - \omega_f$ between the two photonic states and the atomic transition at $\omega_{at}$. Upon insertion of Eqs. (3.8) and (3.9) into the master equation Eq. (3.11) it is immediate to see that the real part of the Lorentzian factor determines the effective emission rate
\[
\Gamma_{em}^{eff} (\omega_{f,f}) = \Gamma_{em} \frac{\Gamma_{pump}^2/4}{(\omega_{at} - \omega_{f,f})^2 + \Gamma_{pump}^2/4}, \tag{3.10}
\]
while the imaginary part introduces a Hamiltonian contribution describing a global frequency shift of the photonic states under the effect of the population-inverted atoms. The width of the Lorentzian is set by the pumping rate $\Gamma_{pump}$, that is by the autocorrelation time $\tau_c = 1/\Gamma_{pump}$ of the atom seen as a frequency-dependent emission bath. Physically, the condition $\Gamma_{em} \ll \Gamma_{pump}$ means that the atomic correlation time is much shorter than the characteristic emission time.

While an extension of our study to the $\Gamma_{loss} \gtrsim \Gamma_{pump}$ regime would only introduce technical complications, entering the $\Gamma_{em} \gtrsim \Gamma_{pump}$ regime is expected to dramatically modify the physics, as a single atom could exchange photons with the cavity at such a fast rate that it has not time to be re-pumped to the excited state between two emission events. As a result, reabsorption processes and Rabi oscillations are possible, which considerably complicate the theoretical description. These issues will be the subject of future investigations.

IV. ONE CAVITY

As a first example of application, we consider the simplest case of a single nonlinear cavity. A special attention will be paid to the stationary state $\rho_{ss}$ of the system for which Eq. (3.11) imposes
\[
0 = -i [H_{ph}, \rho_{ss}] + \mathcal{L}_{loss}(\rho_{ss}) + \mathcal{L}_{em}(\rho_{ss}). \tag{4.1}
\]
In our specific case of a single cavity, the photonic states are labelled by the photon number $N$ and have an energy
\[
\omega_N = N\omega_{cav} + \frac{1}{2}N(N-1)U. \tag{4.2}
\]
Correspondingly, the $N \rightarrow N + 1$ transition has a frequency
\[
\omega_{N+1,N} = \omega_{cav} + NU, \tag{4.3}
\]
and the corresponding photon emission rate is
\[
\Gamma_{em}(\omega_{N,N-1}) = \Gamma_{em,0} \frac{(\Gamma_{pump}/2)^2}{(\omega_{N,N-1} - \omega_{at})^2 + (\Gamma_{pump}/2)^2}. \tag{4.4}
\]

Figure 1: Comparison of the analytical prediction of the photonic model (solid black line) to the numerical solution of the full atom-cavity master equation (open red points). Stationary value of the average number of photons as a function of the photon loss rate $\Gamma_{loss}$ (left) and of the atom-cavity coupling $\Omega_R$ (right). Parameters: $2U/\Gamma_{pump} = 2$, $2\Omega_R/\Gamma_{pump} = 0.02$ (left a) panel; $2U/\Gamma_{pump} = 0.6$, $2\Gamma_{loss}/\Gamma_{pump} = 0.02$ (right b) panel. In all panels, $2S/\Gamma_{pump} = 8$.

A. General features and validation of the method

In the single cavity case, states are fully labelled by the number $N$ of photons and no coherence can exist between states with different $N$. As a result, the stationary density matrix is diagonal in the Fock basis, $\rho_{ss} = \delta_{N,N'}\pi_N$ with the populations $\pi_N$ satisfying
\[
(N + 1)\Gamma_{loss}\pi_{N+1} - (N + 1)\Gamma_{em}(\omega_{N+1,N})\pi_N + N\Gamma_{em}(\omega_{N,N-1})\pi_{N-1} - N\Gamma_{loss}\pi_N = 0, \tag{4.5}
\]
where the two terms on the second line are always zero for $N = 0$. As only states with neighboring $N$ are connected by the emission/loss processes, detailed balance is automatically enforced in the stationary state, which imposes the simple condition on the populations,
\[
(N + 1)\Gamma_{loss}\pi_{N+1} - (N + 1)\Gamma_{em}(\omega_{N+1,N})\pi_N = 0 \tag{4.6}
\]
which is straightforwardly solved in terms of a product,
\[
\pi_N = \pi_0 \prod_{M=0}^{N-1} \frac{\Gamma_{em}(\omega_{M+1,M})}{\Gamma_{loss}} = \left( \frac{\Gamma_{em,0}}{\Gamma_{loss}} \right) \prod_{M=0}^{N-1} \frac{(\Gamma_{pump}/2)^2}{(\omega_{M+1,M} - \omega_{at})^2 + (\Gamma_{pump}/2)^2} \pi_0. \tag{4.7}
\]

Before proceeding, it is useful to compare this analytical prediction to the stationary state of the atom-cavity system, as obtained from a numerical solution of the full master equation Eq. (2.4). For example, in the left panel of Fig. 1B the stationary value for the average photon number is plotted as a function of the photon loss rate $\Gamma_{loss}$. As expected, the purely photonic approach based on the projective method gives very accurate
results as long as the pump rate (i.e. the inverse autocorrelation time of the atomic bath) is much faster than the loss rate \( \Gamma_{\text{loss}} \).

A similar plot of the average photon number as a function of the atom-cavity coupling \( \Omega_R \) is shown in the right panel. Outside the small \( \Omega_R \) regime, the photonic theory tends to overestimate the photon number. This deviation can be explained as the theory assumes the atoms to be always in their excited state ready for emission and neglects the possibility of an atom reabsorbing the emitted photon before being re-pumped to the excited state.

Note that in both panels no constraint needs to be put on the parameters of the free photonic Hamiltonian: the purely photonic theory well matches the numeric prediction also when the parameter \( U \) is plotted in Fig.3. At short times, the value of \( N \) is only possible as a result of a large fluctuation, so it has a very low probability. For \( U \ll \Gamma_{\text{pump}} \), the cavity mode starts close to resonance, one can achieve the regime where the density matrix for the cavity shows a monotonically decreasing thermal occupation law. For strong pumping and/or large detuning, one has

\[
\Gamma_{\text{em},0} \frac{(\Gamma_{\text{pump}}/2)^2}{\delta^2 + (\Gamma_{\text{pump}}/2)^2} < \Gamma_{\text{loss}},
\]

so the density matrix for the cavity shows a monotonically decreasing thermal occupation law. For strong pumping and close to resonance, one can achieve the regime where the emission overcompensates losses and the cavity mode starts being strongly populated:

\[
\Gamma_{\text{em},0} \frac{(\Gamma_{\text{pump}}/2)^2}{\delta^2 + (\Gamma_{\text{pump}}/2)^2} > \Gamma_{\text{loss}}.
\]

The transition between the two regimes is the usual laser threshold, but our purely photonic theory is not able to include the gain saturation mechanism that serves to stabilize laser oscillation. Within our model, the population would in fact show a clearly unphysical monotonic growth for increasing \( N \). A complete description in terms of the full atom-cavity master equation would of course solve this pathology including a gain saturation mechanism according to usual laser theory, but this goes beyond the scope of the present work.

For \( U > 0 \), the situation is much more interesting as the effective transition frequency depends on the number of photons,

\[
\omega_{N+1,N} = \omega_{\text{cav}} + NU \geq \omega_{\text{cav}}.
\]

so the lasing condition

\[
\frac{\Gamma_{\text{em},0}}{\Gamma_{\text{loss}}} \frac{(\Gamma_{\text{pump}}/2)^2}{(\omega_{N+1,N} - \omega_{\text{at}})^2 + (\Gamma_{\text{pump}}/2)^2} \geq 1 \quad (4.12)
\]

can be satisfied (provided \( \Gamma_{\text{em},0} > \Gamma_{\text{loss}} \)) in a finite range of photon numbers only.

This physics is illustrated in Fig2(a). For \( \Gamma_{\text{em},0} < \Gamma_{\text{loss}} \), losses always dominate. For \( \Gamma_{\text{em},0} > \Gamma_{\text{loss}} \), the lasing condition is instead satisfied in a range of frequencies \( [\omega_1, \omega_2] \) around \( \omega_{\text{at}} \). Under the weak nonlinearity condition \( U \ll \Gamma_{\text{pump}} \), the \( [\omega_1, \omega_2] \) range typically contains a large number of transition frequencies \( \omega_{N,N+1} \) at different \( N \).

Three different regimes can then be identified depending on the position of the cavity frequency \( \omega_{\text{cav}} \) with respect to the \( [\omega_1, \omega_2] \) range. (i) If \( \omega_2 \leq \omega_{\text{cav}} \), then the lasing condition is never verified, and the population \( \pi_N \) shown in Fig2(b) is a monotonically decreasing function of \( N \). In this regime, the physics is very similar to the thermal state below threshold.

(ii) If \( \omega_1 \leq \omega_{\text{cav}} \leq \omega_2 \), the population \( \pi_N \) shown in Fig2(c) is an increasing function for small \( N \), then shows a maximum for \( N \approx N' = (\omega_2 - \omega_{\text{cav}})/U \), and finally begins to monotonically decrease for \( N > N' \).

(iii) If \( \omega_{\text{cav}} \leq \omega_1 \), for small \( N \) the population \( \pi_N \) decreases from its initial value \( \pi_0 \) until the nonlinearly shifted frequency enters in the lasing interval for \( N \approx N'' = (\omega_1 - \omega_{\text{cav}})/U \). After this point \( \pi_N \) starts increasing again until it reaches a local maximum at \( N \approx N'' = (\omega_2 - \omega_{\text{cav}})/U \). Finally, for even larger \( N \) it begins to monotonically decrease. An example of this complicated behaviour is shown in Fig2(d). The existence of two well separate local maxima at \( N = 0 \) and \( N \approx N'' \) in the photon number distribution \( \pi_N \) suggests that the incoherently driven nonlinear cavity exhibits a sort of bistable behaviour: when it is prepared at one maximum, it has the tendency to remain in its neighborhood for a macroscopically long time as compared to all microscopic scales of the problem. Switching from one metastable state to the other results is only possible as a result of a large fluctuation, so it has a very low probability.

This feature is clearly visible in the temporal dependence of the delayed two-photon correlation function

\[
g^{(2)}(\tau) = \frac{\langle a^{\dagger}(t) a^{\dagger}(t+\tau) a(t+\tau) a(t) \rangle_{ss}}{\langle a^{\dagger}(t) a(t) \rangle_{ss} \langle a^{\dagger}(t) a(t) \rangle_{ss}} : \quad (4.13)
\]

that is plotted in Fig2. At short times, the value of \( g^{(2)}(\tau) \) is determined by a weighted average of the contribution of the two maxima according to the stationary \( \pi_N \). After a quick transient due to the dynamics around each maximum of \( \pi_N \), the \( g^{(2)}(\tau) \) correlation function slowly decays to its asymptotic value 1 on a very long time-scale mainly set by the switching time from one maximum to the other.

C. Photon number selection in strongly nonlinear cavities

In the opposite limit \( U \gg \Gamma_{\text{pump}} \), the nonlinearity is so large that a change of photon number by a single unity has a sizable effect on the emission rate \( \Gamma_{\text{em}}(\omega_{N,N+1}) \). The physics
is most clear in the regime when the maximum emission rate is large but only a single transition fits within the emission lineshape: these assumptions are equivalent to imposing that $\Gamma_{em,0} \gg 1$ and $\Gamma_{em,0} \frac{\Gamma_pump}{U^2} \ll 1$, with the further condition that the emission is resonant with the $N_0 \rightarrow N_0 + 1$ transition, $\omega_{at} = \omega_{cav} + N_0 U$. As a result, only this last transition is dominated by emission, while all others are dominated by losses.

In terms of the diagrams in Fig. 2, the stationary distribution $\pi_N$ is therefore sharply peaked at two specific values, $N = 0$ and at $N = N_0$. Examples of this physics are illustrated in Fig. 4 depending on the parameters, the relative height of the two peaks can be tuned to different values almost at will. It is however important to note that having a sizable stationary population in the $N = N_0$ state requires quite extreme values
of the parameters as population would naturally tend to accumulate at $N = 0$: this difficulty is exponentially harder for larger $N_0$.

This feature can be easily explained in terms of the asymmetry in the switching mechanisms leading from $N = 0$ to $N = N_0$ and viceversa. The former process requires in fact a sequence of several unlikely emission events from $N = 0$ to $N = N_0 - 1$ as emission is favoured only in the last step. On the other hand, decay from $N = N_0$ occurs as a consequence a single unlikely loss event from $N = N_0 - 1$ to $N = N_0 - 2$: as soon as the system is at $N = N_0 - 2$, it will quickly decay to $N = 0$. The frequency of such an accident can be estimated as follows: the probability that the system in $N = N_0 - 1$ decays to $N = N_0 - 2$ is a factor $\left(\frac{N_0 - 1}{N_0}\right)^{\Gamma_{\text{loss}}} \approx N_0^{\Gamma_{\text{loss}}}$ smaller than the one of being repumped to $N = N_0$. The rate at which the system decays from $N = N_0$ to $N_0 - 1$ is approximately equal to $N_0^{\Gamma_{\text{loss}}}$, which finally gives

$$\Gamma_{\text{acc}} = N_0^{\Gamma_{\text{loss}}} \left(\frac{N_0 - 1}{N_0}\right)^{\Gamma_{\text{loss}}} \ll N_0^{\Gamma_{\text{loss}}}. \quad (4.14)$$

This longer time scale $\tau_{\text{acc}} = \Gamma_{\text{acc}}^{-1}$ is clearly visible in the long tail of the time-dependent $g^{(2)}(t)$ that is plotted in Fig.3. The quick feature at very short times corresponds to the emission rate $\Gamma_{\text{em}}$.

If needed, the characteristic time scale $\tau_{\text{acc}}$ could be further enhanced by adding a second atomic species whose transition frequency is tuned to quickly and selectively emit photons on the $N - 2 \rightarrow N - 1$ transition. In this way, the accident rate can be efficiently reduced to $\Gamma_{\text{acc}} \simeq \Gamma_{\text{loss}}^{(2)} \left(\Gamma_{\text{loss}} / \Gamma_{\text{em},0}\right)^2 \ll \Gamma_{\text{acc}}$. By repeating the mechanism on $k$ transitions, one can suppress the accident rate in a geometrical way to $\Gamma_{\text{acc}}^{(k)} \simeq \Gamma_{\text{loss}}^{(k)} \left(\Gamma_{\text{loss}} / \Gamma_{\text{em},0}\right)^k \ll \Gamma_{\text{acc}}$.

From a slightly different perspective, the slow rate of accidents $\Gamma_{\text{acc}}$ can be exploited to selectively prepare a metastable state with $N = N_0$ photons even in parameter regimes where the $N = 0$ state would be statistically favoured at steady-state. Even if the state will eventually decay to $N = 0$, its
lifetime can be long enough to use it for interesting experiments. This idea removes the necessity of a balance between \( \pi(N) \) and \( \pi(0) \) and so the need for extreme parameters such as the ones used in Fig.4 as a result, the difficulty of creating a (metastable) state of \( N_0 \) photons is roughly independent of \( N_0 \).

The idea to prepare the state with \( N_0 \) photons is to inject a larger number \( N > N_0 \) of photons into the cavity: the system will quickly decay to the \( N = N_0 \) state where the system remains trapped with a lifetime \( \Gamma^{-1}_{acc} \). The efficiency of this idea is illustrated in Fig.6 where we plot the time evolution of the most relevant populations \( \pi_N \). With respect to Fig.4 the chosen parameters are much less extreme. The initially created state with \( N = N_{in} \) photons quickly decays, so that population accumulates into \( N = N_0 \) on a time-scale of the order of \( \Gamma_{loss} \), the eventual decay of the population towards \( N = 0 \) will then occur on a much longer time set by \( \Gamma_{acc} \). It is worth noting that this strategy does not require that the initial preparation be number-selective: it will work equally well if a wide distribution of \( N_{in} \) are generated at the beginning, provided a sizable part of the distribution lies at \( N > N_0 \).

These results show the potential of this novel photon number selection scheme to create Fock states: upon sudden switch-off of the cavity mirrors, one would obtain a quite novel quantum optical wavepacket containing an exact number of photons sharing the same wavefunction.

V. CAVITY ARRAYS

After having unveiled a number of interesting features that occur in the simplest case of a single-cavity, we are now in a position to start attacking the far richer many-cavity case. Throughout this section, we shall make heavy use of the photonic description, which allows to consider bigger systems with a higher number of photons.

A. Validation of the method

As a first step, we have further validated the photonic description by comparing its predictions with the numerical results for the full atom-cavity master equation in a two cavity case. An example is shown in Fig.7 as in the single cavity case, the agreement is excellent at large \( \Gamma_{pump} \) and gets deteriorated when \( \Gamma_{pump} \) is decreased to values comparable to \( \Gamma_{loss} \) [panel (a)]. The situation is even more favourable in panel (b), where the deviations that are expected for larger \( \Omega_R \) are suppressed by the strong nonlinearity. Based on these numerical results, we are reassured of the validity of the analytical approximations underlying our the photonic approach: all discussions in the following will be based on this latter approach.
For a single cavity, the stationary state is immediately obtained as a function of the photon loss rate $\Gamma_{\text{loss}}$ (left) of the atom-cavity coupling $\Omega R$ (right). Parameters: $2U/\Gamma_{\text{pump}} = 7$, $2\Omega R/\Gamma_{\text{pump}} = 0.02$, (left a) panel); $2U/\Gamma_{\text{pump}} = 28$, $2\Gamma_{\text{loss}}/\Gamma_{\text{pump}} = 0.002$ (right b) panel). In all panels, $2J/\Gamma_{\text{pump}} = 2$ and $\delta = 0$.

**B. Markovian regime**

We begin by considering the Markovian limit of the theory, which is recovered for $\Gamma_{\text{pump}} \to \infty$, i.e., for a frequency-independent gain. In this case, the emission term of the master equation for photons Eq. (3.3) reduces to the usual Lindblad form

$$\mathcal{L}_{\text{em}} = \frac{\Gamma_{\text{em}}}{2} \sum_{i=1}^{k} \left[ 2a_i^{+}\rho a_i - a_i a_i^{+}\rho - \rho a_i a_i^{+} \right].$$

(5.1)

For a single cavity, the stationary state is immediately obtained as

$$\pi_N = \frac{1}{1 - \frac{\Gamma_{\text{em}}}{\Gamma_{\text{loss}}}} \left( \frac{\Gamma_{\text{em}}}{\Gamma_{\text{loss}}} \right)^N,$$

(5.2)

a necessary condition for stability for this system is that $\Gamma_{\text{em}} < \Gamma_{\text{loss}}$. If $\Gamma_{\text{em}} > \Gamma_{\text{loss}}$, amplification overwhelms losses and the system displays a laser instability: while a correct description of gain saturation is beyond the photonic model, the full atom-cavity model recovers for this model the standard laser operation.

For larger arrays of $k$ sites, a straightforward calculation shows that in the Markovian limit the stationary matrix keeps a structureless form,

$$\rho_\infty = \sum_N \pi_N \mathcal{I}_N,$$

(5.3)

with

$$\pi_N = \frac{1}{\sum_M D_M \left( \frac{\Gamma_{\text{em}}}{\Gamma_{\text{loss}}} \right)^M} \left( \frac{\Gamma_{\text{em}}}{\Gamma_{\text{loss}}} \right)^N.$$

(5.4)

Here, $D_N = \frac{(N + k - 1)!}{(k - 1)! N!}$ is the dimension of the Hilbert subspace with a total number of photons equal to $N$ and $\mathcal{I}_N$ is the projector over this subspace.

This result shows that independently of the number of cavities and the details of the Hamiltonian, in the Markovian limit the density matrix in the stationary state corresponds to an effective Grand-Canonical ensemble at infinite temperature $\beta = 0$ with a fugacity $z = e^{\beta \mu} = \Gamma_{\text{em}}/\Gamma_{\text{loss}}$ determined by the pumping and loss conditions only: All states are equally populated and the system can hardly display any interesting physics. In the next subsection we shall see how inclusion of non-Markovian effects stemming from the frequency-dependence of the emission can radically change the picture giving rise to interesting physics.

**C. Effective Grand-Canonical distribution in a weakly non-Markovian and secular regime**

In this section we consider a case where all relevant transitions adding one photon have a narrow distribution around the bare cavity frequency, $|\omega_{f f'} - \omega_{\text{cav}}| \ll \Gamma_{\text{pump}}$. We also assume a secular limit where $U, J \gg \Gamma_{\text{em}}, \Gamma_{\text{loss}}$, so that the non-diagonal terms of the density matrix oscillate at a fast rate and are thus effectively decoupled from the (slowly varying) populations. In this limit, we can safely assume that all coherences vanish and we can restrict our attention to the populations.

Along these lines, the transfer rate on the $|f'\rangle \to |f\rangle$ transition where one photon is lost from $N + 1$ to $N$ has a frequency-independent form

$$T_{f \to f'} = \Gamma_{\text{loss}} \langle |f'|\ a\ |f\rangle^2,$$

(5.5)

while the reverse emission process depends on the detunings $\Delta_{f f'} = \omega_{f f'} - \omega_{\text{cav}}$ and $\delta = \omega_{\text{cav}} - \omega_{\text{at}}$ as

$$T_{f \to f'} = \Gamma_{\text{em},0} \left| \langle f'\ |a\ |f\rangle \right|^2 \frac{\Gamma_{\text{pump}}^2}{(\Delta_{f f'} + \delta)^2 + \frac{\Gamma_{\text{pump}}^2}{4}} \simeq \Gamma_{\text{em},0} \left| \langle f'\ |a\ |f\rangle \right|^2 \left[ 1 - \beta \Delta_{f f'} + \mathcal{O} \left( \Delta_{f f'}^2 \right) \right],$$

(5.6)

with

$$\Gamma_{\text{em},0} = \frac{\left( \Gamma_{\text{pump}}/2 \right)^2}{\delta^2 + \left( \Gamma_{\text{pump}}/2 \right)^2},$$

(5.7)

$$\beta = \frac{2\delta}{\delta^2 + \left( \Gamma_{\text{pump}}/2 \right)^2}.$$  

(5.8)

In this terms, the weakly non-Markovian regime is characterized by $|\beta \Delta_{f f'}| \ll 1$. Without loss of accuracy, we can then replace the square bracket in Eq. (5.6) by an exponential

$$1 - \beta \Delta_{f f'} \simeq e^{-\beta \Delta_{f f'}},$$

(5.9)

which immediately leads to a Grand-Canonical form of the stationary density matrix

$$\rho_\infty = \frac{1}{z} e^{\beta N \mu} e^{-\beta H},$$

(5.10)
pared to the pumping of photons allowed in the system to induces global measurable and an effective temperature $k_B T = 1/\beta$. Even if each transition involves a small deviation from the bare cavity frequency $\omega_{cav}$, the cumulative effect of many deviations can become important compared to $1/\beta$ and make the stationary distribution strongly non-trivial. From Eq.(5.8), it is interesting to note that both positive and negative temperature configurations can be obtained just by tuning the peak emission frequency $\omega_{at}$ below or above the bare cavity frequency $\omega_{cav}$.

As expected for a thermal-like distribution, detailed balance between eigenstates is satisfied

$$T_{f' \rightarrow f} = T_{f \rightarrow f'} = \frac{|\langle f' | a | f \rangle|^2}{\beta \omega_{f'} - \beta \omega_f} \rho_a$$

but it is crucial to keep in mind that this thermal-like distribution does not arises from any real thermalization process, but is a consequence of the specific form chosen for the pumping and dissipation. The relation between this prediction and the experimental observations of a thermal Boltzmann-like tail in the momentum distribution observed in recent photon \cite{36} and polariton \cite{18,37} Bose-Einstein condensation experiments will be the subject of future work.

This result can be numerically tested on a two cavity system with small non linearity $U$ and tunnel coupling $J$ compared to the pumping $\Gamma_{pump}$, but with large enough number of photons allowed in the system to induces global measurable non linear effects on photon number and correlation functions. The results of this comparison are shown in Fig.8: excellent agreement is found in both the average photon number and the first-order coherence between the cavities by the exact resolution of the photonic master equation and the grand canonical ensemble ansatz.

![Figure 8: Two cavity system with nonlinearity $U$ and tunnel coupling $J$ small as compared to the pumping $\Gamma_{pump}$. In red dots, exact resolution of the photonic master equation, and in black solid line the grand canonical ensemble ansatz. The left panel shows the average quantity of photons $n_1 = \langle c_1^\dagger c_1 \rangle$ in the first cavity, the right panel shows the spatial coherence $g_{12}^\dagger = \langle c_1^\dagger c_2 \rangle / \langle c_1^\dagger c_1 \rangle$ as a function of the non linearity $2U/\Gamma_{pump}$. Parameters : $2J/\Gamma_{pump} = 0.02$, $2\Gamma_{loss}/\Gamma_{pump} = 0.002$, $2\Gamma_{em}/\Gamma_{pump} = 0.0014$, $2\delta/\Gamma_{pump} = 0.6$](image1)

![Figure 9: Dependence of the relative quantum coherence between two arbitrarily chosen two-photon eigenstates $\rho_{ij}/\sqrt{\rho_{ii}\rho_{jj}}$ on the atomic autocorrelation time (scaling as $1/\Gamma_{pump}$). As expected this coherence vanishes in $1/\Gamma_{pump}$ in the Markovian limit $1/\Gamma_{pump} \rightarrow 0$. The value above 1 for large $1/\Gamma_{pump}$ signals breakdown of positivity of the density matrix as we move out of the validity regime of this master equation. Parameters: $J/\Gamma_{loss} = 1$, $\Gamma_{em}/\Gamma_{loss} = 0.5$, $\delta = -\Gamma_{loss}, U/\Gamma_{loss} = 2$.](image2)
with
\[
\delta \mathcal{M}[\rho] = \frac{\Gamma_{em,0}}{2} \sum_{i=1}^{k} \left[ \delta a_{i}^{\dagger} \rho a_{i} + a_{i}^{\dagger} \rho \delta a_{i} - a_{i} \delta a_{i}^{\dagger} \rho - \rho \delta a_{i} a_{i}^{\dagger} \right],
\]
(5.14)
with
\[
\tilde{\delta} a_{i}^{\dagger} = \delta a_{i}^{\dagger} + \tilde{\delta} a_{i},
\]
(5.15)
and
\[
\langle f'| \tilde{\delta} a_{i}^{\dagger} | f \rangle = e^{-\beta(\Delta \omega_{ff'} - \omega_{em})} \langle f'| \delta a_{i}^{\dagger} | f \rangle.
\]
(5.16)
we deduce that
\[
\langle f'| \delta a_{i}^{\dagger} | f \rangle = \frac{\Gamma_{pump}}{\Gamma_{pump} \to \infty} \langle f'| \delta a_{i}^{\dagger} | f \rangle \left( -\frac{(\omega_{ff'} - \omega_{em})}{\Gamma_{pump}} + \mathcal{O} \left( \frac{\Delta f_{pump}}{\Gamma_{pump}} \right)^{2} \right).
\]
(5.17)
We can easily show then in a very similar way to the demonstration made in appendix C for Markovian case, that
\[
\mathcal{M}_{0}(e^{\beta N \mu}e^{-\beta H}) = 0,
\]
(5.18)
so that the grand canonical distribution is a steady state of this modified \( \mathcal{M}_{0} \) operator. Also, \( \delta \mathcal{M} \) vanishes in the Markovian limit proportionally to \( 1/\Gamma_{pump} \), so we can calculate the correction of the steady state at lowest order in \( \delta \mathcal{M} \).

Expanding the steady state to next order in \( 1/\Gamma_{pump} \), we see easily that populations are perturbed only to second order in \( \beta \Delta \omega_{ff'} \), because the first order corrections in eq. (5.17) are purely imaginary. In our Markovian limit, these corrections then vanish even if we perform simultaneously the Markovian and thermodynamic limit.

Secondly, coherences (which are exactly zero in the Markovian case) should be then proportional to \( i(\omega_{ff'} - \omega_{em})/\Gamma_{pump} \). However, we have shown in appendix C that the linear contribution to coherences vanishes when we sum over all sites of the system. We conclude thus that in the weakly non Markovian limit, coherences between eigenstates of the hamiltonian are quadratic in \( 1/\Gamma_{pump} \), and are therefore very small, even out of the secular approximation. To support this analytical argument, in Fig.9 we have shown the dependence of the coherence between an arbitrary pair of two-photon states on the autocorrelation time. We also represented the error in population of an arbitrary eigenstate, between the true steady state, and the grand canonical distribution. Both scale as \( (1/\Gamma_{pump})^{2} \).

D. Two cavities with strong non linearity: Mott-insulator physics

As a final example of application of our concepts, in this section we study the case of two strongly nonlinear cavities with \( U \gg \Gamma_{pump} \); extending the photon-number selectivity idea to the many-cavity case, we look for states that resemble a Mott insulator. As in the single cavity case, strong pumping \( \Gamma_{em} \gg \Gamma_{loss} \) favours larger occupations of sites, while the nonlinearity \( U \gg \Gamma_{pump} \) puts an upper bound to the occupation: the result is a steady-state with a well-defined number of photons per cavity.

Numerical calculations based on the photonic master equation are shown in Fig.10 panels a-c, black lines) for a Mott state with one photon per site (zero detuning between the atoms and the cavity): for high pumping rate and strong non linearity, signatures of the desired Mott state are visible in the steady-state average number of photons (panel a) that tends to 1 for a strong nonlinearity \( U \), in the probability of double occupancy (panel b) that tends to 0, and in the one-body coherence between the two sites (panel c), that tends to 0. While these results are a strong evidence of \( N = 1 \) Mott state, a similar calculation for larger \( N \geq 2 \) Mott states are much more tricky because of metastability issues and the Mott state typically has a finite lifetime.

Red dashed lines in panels a-c of Fig.10 show the same simulation for a weaker emission rate, which allows to consider weaker values of the nonlinearity without increasing too much the photon number. In particular, in panel c, the non-negligible value of \( 2J/\Gamma_{pump} \) is responsible for significant spatial coherence between the two sites: this quantity a maximum \( g_{12}^{(1)} = 0.31 \) for an interaction strength \( 2U/\Gamma_{pump} \approx 0.16 \) of the same order of magnitude as the tunnel coupling \( 2J/\Gamma_{pump} \approx 0.2 \). The appearance of this coherence can be understood as follows: on one hand, without any tunneling \( (J = 0) \), all the dynamics is local and we do not expect any spatial coherence. On the other hand, in the absence of interactions \( (U = 0) \), for zero detuning, symmetric and anti-symmetric states are equally close to resonance (albeit with opposite detuning) and then equally populated, so there should not be any coherence either. However in presence of both tunnelling and small interactions \( (J \neq 0, U \neq 0, U \ll J) \), to first order in perturbation theory, the energy of all eigenstates (symmetric/anti-symmetric states with various photon numbers) is shifted in the upward direction. As a result, symmetric states, which are below the resonance, get closer to resonance and become more populated than the anti-symmetric ones, which get farther to the resonance and are thus depleted. This induces an overall positive coherence between the two sites. Even though the nonlinearity is only active for states with at least two photons, it is interesting to note that even the one-particle antisymmetric state is less populated than the symmetric one. This is due to the photonic decay from above-lying eigenstates with more than one photon, which preferentially decay into the symmetric state. So, no coherence is expected in both limiting cases of purely interacting and non interacting photons, we expect the coherence reaches a maximum when interactions and tunnelling are of the same magnitude. To visually illustrate this physics, in panel d of Fig.10 we plotted the energy, the spatial coherence and the occupancy at steady state of the different eigenstates of the hamiltonian at the point of maximum coherence.
VI. CONCLUSIONS

In this work we have proposed and characterized a novel scheme to generate strongly correlated states of light in strongly nonlinear cavity arrays. Photons are incoherently injected in the cavities using population-inverted two-level atoms, which preferentially emit photons around their resonance frequency. The resulting frequency-dependence of the gain will be the key element to select and stabilize the desired quantum state. A manageable theoretical description of the system is obtained using projective methods, which allow to eliminate the atomic degrees of freedom and describe the non-Markovian photonic dynamics in terms a generalized master equation.

The efficiency of the our pumping scheme to generate specific quantum states is first validated on a single-cavity system: for weak nonlinearities, a novel mechanism for optical bistability is found. For strong nonlinearities, Fock states with a well-defined photon number can be generated with small number fluctuations.

In the general many cavity case, in the weakly non-Markovian case the steady-state of the system recovers a Grand-Canonical distribution with an effective chemical potential determined by the pumping strength and an effective inverse temperature proportional to the non-Markovianity: This very general results may have application to explain apparent thermalization in recent photon and polariton condensation experiments.
Finally, the power of a frequency-dependent pumping to generate strongly correlated states of light is illustrated in the case of a strongly nonlinear two-cavity system which, in the strongly non-Markovian regime, can be driven into a Mott-insulator state. A general study of the potential and of the limitations of the frequency-dependent gain to generate generic strongly correlated states with many photons will be the subject of future work.

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Appendix A: Projective methods

In this Appendix, we give more details on the derivation of the photonic master equation (31). Starting from the full atom-cavity master equation (2), we show how for a sufficiently small atom-cavity coupling $\Omega_0$ the atomic degrees of freedom can be eliminated. The frequency-dependence of the atomic amplification is then accounted for as a modified Lindblad term (33). Our treatment is based on the discussion in the textbook [40].

1. General formalism

We consider a quantum system which undergoes dissipative processes. Since, it is not isolated, in general it can not be described by a wave function but by its density matrix $\rho$. This matrix follows the following evolution:

$$\partial_t \rho = \mathcal{L}(\rho(t)), \quad (A1)$$

where $\mathcal{L}$ is some generalized super operator for the density matrix. Given an arbitrary initial density matrix $\rho(t_0)$, the density matrix $\rho$ at generic time $t$ is equal to $\rho(t) = e^{\mathcal{L}(t-t_0)} \rho(t_0)$.

Now we are only interested in some part of the density matrix, which can represent some subsystem. This can be described by a projection operation on the density matrix $\mathcal{P} \rho$. We call $Q = 1 - \mathcal{P}$ the complementary projector. We decompose the Lindblad operator $\mathcal{L}$ in two parts $\mathcal{L}_0$ and $\delta \mathcal{L}$ such that:

$$\begin{cases}
\mathcal{L} = \mathcal{L}_0 + \delta \mathcal{L} \\
\mathcal{P} \mathcal{L}_0 Q = Q \mathcal{L}_0 \mathcal{P} = 0 \\
\mathcal{P} \delta \mathcal{L} \mathcal{P} = 0.
\end{cases} \quad (A2)$$

Such a decomposition is always possible.

Then we define the density matrix and superoperators in the generalised interaction picture for superoperators with respect to the evolution described by the free $\mathcal{L}_0$ and the initial time $t_0$:

$$\begin{cases}
\hat{\rho}(t) = e^{-\mathcal{L}_0(t-t_0)} \rho(t) \\
\hat{\mathcal{A}}(t) = e^{-\mathcal{L}_0(t-t_0)} \mathcal{A} e^{\mathcal{L}_0(t-t_0)}.
\end{cases} \quad (A3)$$

where $\mathcal{A}$ is some superoperator acting on the density matrix.

As discussed in [41], we can get an exact closed master equation for the projected density matrix in the interaction picture

$$\partial_t \mathcal{P} \hat{\rho}(t) = \int_{t_0}^t dt' \Sigma(t, t') \mathcal{P} \hat{\rho}(t'), \quad (A4)$$

which translates to

$$\partial_t \mathcal{P} \rho(t) = \mathcal{L}_0(\rho(t)) + \int_{t_0}^t dt' \Sigma(t - t') \mathcal{P} \rho(t') \quad (A5)$$

in the Schrodinger picture. The self energy operator $\Sigma$ is defined as:

$$\Sigma(t, t') = \sum_{n=2}^\infty \int_{t'}^{t_1} dt_1 \cdots \int_{t'}^{t_{n-1}} dt_{n-1} \mathcal{P} \delta \hat{\mathcal{L}}(t) \mathcal{Q} \delta \hat{\mathcal{L}}(t_1) \mathcal{Q} \delta \hat{\mathcal{L}}(t_2) \cdots \mathcal{Q} \delta \hat{\mathcal{L}}(t_{n-1}) \delta \hat{\mathcal{L}}(t') \mathcal{P} \quad (A6)$$

and

$$\hat{\Sigma}(t - t') = e^{\mathcal{L}_0(t-t_0)} \Sigma(t, t') e^{-\mathcal{L}_0(t-t_0)}. \quad (A7)$$

It results from the coherent sum over the processes leaving from $\mathcal{P}$, remaining in $\mathcal{Q}$ and then coming back finally to $\mathcal{P}$ (in the Feynman vocabulary, it is the sum over all irreducible diagrams).

We call $\tau_c = 1/\Delta \omega$ the characteristic decay time / inverse linewidth for the self energy, which corresponds in general to the correlation time of the bath, and we define $\Gamma = \sum \tau_c = \int_{t_0}^\infty dt \Sigma(t, t_0)$ as an estimation of the intensity of dissipative process. We put ourselves in the regimes in which, with respect to these dissipative processes, the bath has a short memory, ie $\Gamma \ll \Delta \omega$. In that regime the density matrix in the interaction picture is almost constant over that time $\tau_c$. Furthermore, if $t - t_0 \gg \tau_c$ then the integral in eq (A3) can be extended from $- \infty$ to $t$. From this equation and from (A4), we get an equation of evolution for the density matrix which is local in time:

$$\partial_t \mathcal{P} \hat{\rho}(t) = \int_0^\infty dt \Sigma(t, t - \tau) \mathcal{P} \hat{\rho}(t)$$

$$= \int_0^\infty d\tau \left[ e^{-\mathcal{L}(t-t_0)} \Sigma(0, -\tau) e^{\mathcal{L}(t-t_0)} \right] \mathcal{P} \hat{\rho}(t)$$

$$= e^{-\mathcal{L}(t-t_0)} \int_0^\infty d\tau \Sigma(0, -\tau) \mathcal{P} \rho(t). \quad (A8)$$

In the Schrodinger picture this gives the time-local master equation:

$$\partial_t \mathcal{P} \rho(t) = \left[ \mathcal{L}_0 + \int_0^\infty d\tau \Sigma(0, -\tau) \right] \mathcal{P} \rho(t) = \mathcal{L}_{eff} \mathcal{P} \rho(t). \quad (A9)$$
with
\[ \mathcal{L}_{\text{eff}} = \mathcal{L}_0 + \int_0^\infty d\tau \Sigma(0, -\tau). \]  

(A10)

It is worth stressing that while the bath is Markovian with respect to dissipative processes induced by the perturbation \( \int_0^\infty d\tau \Sigma(0, -\tau) \) (which accounts for photonic emission in our system), no Markovian approximation has been made with respect to the dynamics due to \( \mathcal{L}_0 \) (in particular the photonic hamiltonian dynamics), which can still be fast.

2. Application to the array of cavities

a. Preliminary calculations

With the notation from section II, we choose the projectors in the form:
\[ \mathcal{P} \rho = \left\langle e_1^{(1)} e_2^{(1)} e_3^{(1)} \ldots \left| e_1^{(1)} e_2^{(1)} e_3^{(1)} \ldots \right\rangle \otimes \mathcal{T}_{\text{rat}}(\rho), \right. \]

(A11)

where we have performed a partial trace over the atoms, and then make the tensor product of the density matrix and the atomic density matrix with all atoms in the excited state. This projection operation gives us direct access to the photonic density matrix. With the notation of the previous section we have:
\[ \mathcal{L}(\rho) = \frac{1}{i} \left[ \mathcal{H}_{\text{ph}} + \mathcal{H}_{\text{at}} + \mathcal{H}_I, \rho \right] + \mathcal{L}_{\text{diss}}(\rho), \]  

(A12)

with
\[ \mathcal{L}_{\text{diss}} = \mathcal{L}_{\text{pump, at}} + \mathcal{L}_{\text{loss, cav}}. \]  

(A13)

We decompose \( \mathcal{L} \) in two contributions. The first one is:
\[ \mathcal{L}_0(\rho) = -i \mathcal{H}_0(\rho) + \mathcal{L}_{\text{loss, cav}}(\rho) \]
\[ - \frac{\Gamma_{\text{pump}}}{2} \sum_{i=1}^{N_{\text{at}}} \sum_{l=1}^{N_{\text{at}}} \left[ \sigma_i^{-(l)} \sigma_i^{+(l)} \rho + \rho \sigma_i^{-(l)} \sigma_i^{+(l)} \right] \]
\[ - \mathcal{P} \left( \sigma_i^{-(l)} \sigma_i^{+(l)} \rho + \rho \sigma_i^{-(l)} \sigma_i^{+(l)} \right) Q, \]  

(A14)

which verifies the condition (A2). The last term in this expression comes from the fact that the pumping term does not verify this condition: as a result, we have to remove the part unixed by projector and put it in the other operator:
\[ \delta \mathcal{L}(\rho) = -i \mathcal{H}_I(\rho) + \frac{\Gamma_{\text{pump}}}{2} \sum_{i=1}^{N_{\text{at}}} \sum_{l=1}^{N_{\text{at}}} \left[ 2 \sigma_i^{+(l)} \rho \sigma_i^{-(l)} \right] \]
\[ - \mathcal{P} \left( \sigma_i^{+(l)} \rho \sigma_i^{-(l)} \right) Q. \]  

(A15)

These two operators then satisfy to the conditions (A2), and we can apply the projection method to get the evolution of \( \mathcal{P} \rho(t) \), i.e., of \( \mathcal{T}_{\text{rat}}(\rho)(t) \). As we are interested in the regime in which \( \Gamma_{\text{pump}} \gg \Omega_R, \Gamma_{\text{loss}} \), we will compute the self energy at the lowest non zero order of these two parameters. Since \( \Gamma_{\text{loss}} \) quantify photonic losses, we will approximate the photonic dynamics as being a Hamiltonian one during the time while the atom is reinjected in the excited state. To this order of precision, the calculation for one cavity is easily generalizable to \( k \) cavities, thus we will restrict for simplicity to the case of one cavity. We have to calculate superoperators for the density matrix in the interaction picture. On a short time, we neglect the effect of photon losses, so the evolution of the operators is only given by
\[ -i \mathcal{H}_0 - \frac{\Gamma_{\text{pump}}}{2} \left[ \sigma_- + \rho \sigma_- \sigma_+ \right] = -i(\mathcal{H}_0 - \mathcal{H}_I)(\rho) \]
which is a non-hermitian hamiltonian operator.

We start with an operator \( \mathcal{A}_L \) consisting in multiplying the density matrix by a matrix \( A \) on the left:
\[ e^{-\mathcal{L}_0 \tau} \mathcal{A}_L e^{\mathcal{L}_0 \tau}(\rho) = \left. e^{-\mathcal{L}_0 \tau} \right| \mathcal{A} \cdot e^{\mathcal{L}_0 \tau}(\rho) \right|_{\mathcal{L}} \]
\[ = e^{-i\mathcal{H}_0(\rho - \mathcal{H}_0')\tau} \cdot \mathcal{A} \cdot e^{-i\mathcal{H}_0 - \mathcal{H}' \tau} \rho \]
\[ = e^{i\mathcal{H}_0(\rho - \mathcal{H}_0')\tau} \mathcal{A} e^{i\mathcal{H}_0 - \mathcal{H}' \tau} \rho. \]  

(A16)

Thus with have:
\[ e^{-\mathcal{L}_0 \tau} \mathcal{A}_L e^{\mathcal{L}_0 \tau} = \left( e^{i\mathcal{H}_0(\rho - \mathcal{H}_0')\tau} \right) \cdot \mathcal{A} \cdot \left( e^{i\mathcal{H}_0 - \mathcal{H}' \tau} \right) \rho. \]  

(A17)

In the same way on the right:
\[ e^{-\mathcal{L}_0 \tau} \mathcal{A}_R e^{\mathcal{L}_0 \tau} = \left( e^{i\mathcal{H}_0 - \mathcal{H}' \tau} \right) \cdot \mathcal{A} \cdot \left( e^{i\mathcal{H}_0(\rho - \mathcal{H}_0')\tau} \right) \rho. \]  

(A18)

We see that the non hermitian part changes sign when we pass from left to right operators.

We apply this to useful operators:
\[ \begin{cases} a_L(t) = (a(t))_L \\ a_R(t) = (a(t))_R \\ a_L^+(t) = (a^+(t))_L \\ a_R^+(t) = (a^+(t))_R \\ \sigma^+_L(t) = e^{i(\omega_{at} - \Gamma_{\text{pump}}) t} \sigma^+_L \\ \sigma^+_R(t) = e^{i(\omega_{at} + \Gamma_{\text{pump}}) t} \sigma^+_R \\ \sigma^-_L(t) = e^{-(\omega_{at} - \Gamma_{\text{pump}}) t} \sigma^-_L \\ \sigma^-_R(t) = e^{-i(\omega_{at} + \Gamma_{\text{pump}}) t} \sigma^-_R \end{cases} \]  

(A19)

with
\[ \begin{cases} a(t) = e^{i\mathcal{H}_p(\mathcal{T}_{\text{rat}}(\rho))(t)} \\ a^+_L(t) = e^{i\mathcal{H}_p(\mathcal{T}_{\text{rat}}(\rho))(t)} a^+_L \end{cases} \]  

(A20)

b. Self energy calculation

We are going to calculate the self energy to the lowest order possible, in \( \Omega_R \).
We have
\[ \delta \mathcal{L} = \mathcal{L}_{\text{pump}} - i(H^+ + H^-) + i(H^+ + H^-)R + H'^n, \] (A21)
with
\[ \begin{aligned}
\mathcal{L}_{\text{pump}}(\rho) &= \Gamma_{\text{pump}} \sigma^+ \rho \sigma^- \\
H^+ &= \Omega_R \sigma^+ a \\
H^- &= \Omega_R \sigma^- a^\dagger \\
H'^n(\rho) &= -\frac{\Gamma_{\text{pump}}}{2} \mathcal{P} [\sigma_{-\sigma} + \rho + \rho \sigma_{-\sigma}] \mathcal{Q}.
\end{aligned} \] (A22)

First we have \( \mathcal{L}_{\text{pump}} \mathcal{P} = H'^n \mathcal{P} = H'_L \mathcal{P} = H'_R \mathcal{P} = 0 \), so
starting from a projected state, we have to start with \( H'_L \) or \( H'_R \). In fact to the lowest order the non zero contributions to the self energy are:
\[ \begin{aligned}
A &= \mathcal{P} (-i) H'_L(t) (-i) H'_L(t') \mathcal{P} \\
B &= \mathcal{P} i H'_R(t) H'_R(t') \mathcal{P} \\
C &= \mathcal{P} i H'_R(t) (-i) H'_L(t') \mathcal{P} \\
D &= \mathcal{P} (-i) H'_L(t) H'_R(t') \mathcal{P} \\
E &= \int_0^t dt' \mathcal{P} \mathcal{L}_{\text{pump}}(t) \tilde{H}'_L(t') \mathcal{P} \\
F &= \int_0^t dt' \mathcal{P} \mathcal{L}_{\text{pump}}(t) \mathcal{Q} (-i) H'_L(t') \mathcal{P} \\
G &= \mathcal{P} H'^n(t) \tilde{H}'_L(t) (-i) H'_R(t') \mathcal{P} \\
H &= \mathcal{P} H'^n(t) \mathcal{Q} (-i) H'_L(t') H'_R(t') \mathcal{P},
\end{aligned} \] (A23)
with
\[ \Sigma(t, t') = A + B + C + D + E + F + G + H. \] (A24)

We then calculate the different processes
\[ \begin{aligned}
A(\mathcal{P} \rho) &= -\Omega_R^2 e^{i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} a(t) a^\dagger(t') \mathcal{P} \rho \\
B(\mathcal{P} \rho) &= -\Omega_R^2 e^{-i(\omega_{at} + \Gamma_{\text{pump}}/2)(t-t')} \mathcal{P} \rho a(t) a^\dagger(t) \\
C(\mathcal{P} \rho) &= \Omega_R^2 e^{i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} \mathcal{P} \rho a(t) a^\dagger(t) \\
D(\mathcal{P} \rho) &= \Omega_R^2 e^{-i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} \mathcal{P} \rho a(t) a^\dagger(t) \\
E(\mathcal{P} \rho) &= \Gamma_{\text{pump}} \Omega_R^2 \int_0^t dt' e^{-\Gamma_{\text{pump}}(t-t')} e^{i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} a^\dagger(t') \mathcal{P} \rho a(t) \\
F(\mathcal{P} \rho) &= \Gamma_{\text{pump}} \Omega_R^2 \int_0^t dt' e^{-\Gamma_{\text{pump}}(t-t')} a^\dagger(t') \mathcal{P} \rho a(t) \\
G(\mathcal{P} \rho) &= -\Gamma_{\text{pump}} \Omega_R^2 \int_0^t dt' e^{i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} a^\dagger(t') \mathcal{P} \rho a(t) \\
H(\mathcal{P} \rho) &= -\Gamma_{\text{pump}} \Omega_R^2 \int_0^t dt' e^{i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} a^\dagger(t') \mathcal{P} \rho a(t).
\end{aligned} \] (A25)

\[ \begin{aligned}
\langle f | a^\dagger f' \rangle &= \frac{\Gamma_{\text{pump}}}{\Gamma_{\text{pump}}^2 + i(\omega_{at} - \omega_{ff})} \langle f | a^\dagger | f' \rangle \\
\langle f' | a f \rangle &= \frac{\Gamma_{\text{pump}}}{\Gamma_{\text{pump}}^2 + i(\omega_{at} - \omega_{ff})} \langle f' | a | f \rangle.
\end{aligned} \] (A29)

\[ \partial_t \rho_{ph} = -i [H_{ph}, \rho_{ph}] + \mathcal{L}'(\rho_{ph}) \] (A27)

At lowest order in \( \Omega_R \), in the interaction picture, the density matrix is constant in the convolution product: \( \dot{\rho}(t-\tau) = \hat{\rho}(t) \), i.e. \( e^{\mathcal{E}_0 \tau} \rho(t-\tau) = \rho(t) \). Making the trace over the bath we get
\[ \begin{aligned}
\partial_t \rho_{ph} &= -i [H_{ph}, \rho_{ph}] + \mathcal{L}'(\rho_{ph}) \\
&= +\Omega_R^2 \int_0^\infty dt e^{i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} a^\dagger(-\tau) \rho_{ph}(t-\tau) a \\
&+\Omega_R^2 \int_0^\infty dt e^{i(\omega_{at} + \Gamma_{\text{pump}}/2)(t-t')} a \rho_{ph}(t-\tau) a \\
&+\Omega_R^2 \int_0^\infty dt e^{i(\omega_{at} - \Gamma_{\text{pump}}/2)(t-t')} a a^\dagger(-\tau) \rho_{ph}(t-\tau) a \\
&+\Omega_R^2 \int_0^\infty dt e^{i(\omega_{at} + \Gamma_{\text{pump}}/2)(t-t')} \rho_{ph}(t-\tau) a(-\tau) a^\dagger,
\end{aligned} \]
then we can perform completely the integral and we get our final form for the non Markovian master equation, which is local in time:
\[ \partial_t \rho = -i [H_{ph}, \rho_{ph}] + \Gamma_{\text{loss}} \left[ 2a a^\dagger - a^\dagger a - \rho a^\dagger a \right] \] (A28)
The non-Markovianity comes from the emission term, which is energy dependant. For various cavities the reasoning is exactly the same and we get the multicavity master equation:

$$\partial_t \rho = -i [H_{ph}, \rho_{ph}] + \frac{\Gamma \text{loss}}{2} \sum_{i=1}^{k} \left[ 2a_i \rho a_i^\dagger - a_i^\dagger a_i \rho - \rho a_i^\dagger a_i \right]$$

with

$$\langle f \mid \hat{a}_i \mid f' \rangle = \frac{\Gamma_{\text{pump}}/2}{i(\omega_{at} - \omega_{f,f}) + \Gamma_{\text{pump}}/2} \langle f \mid a_i \mid f' \rangle$$

$$\langle f' \mid \hat{a}_i^\dagger \mid f \rangle = -i(\omega_{at} - \omega_{f,f}) + \Gamma_{\text{pump}}/2 \langle f' \mid a_i^\dagger \mid f \rangle.$$  \hspace{1cm} (A30)

**Appendix B: Exact stationary solution for Markovian case**

We are looking for the steady state for the quantum dynamical process:

$$\partial_t \rho = -i [H, \rho(t)] + \mathcal{L}_{\text{loss}} + \mathcal{L}_{\text{em}},$$  \hspace{1cm} (B1)

with the Markovian Lindblad operators:

$$\mathcal{L}_{\text{loss}} = \frac{\Gamma_{\text{loss}}}{2} \sum_{i=1}^{k} \left[ 2a_i \rho a_i^\dagger - a_i^\dagger a_i \rho - \rho a_i^\dagger a_i \right],$$  \hspace{1cm} (B2)

$$\mathcal{L}_{\text{em}} = \frac{\Gamma_{\text{em}}}{2} \sum_{i=1}^{k} \left[ 2a_i^\dagger \rho a_i - a_i \rho a_i^\dagger - \rho a_i a_i^\dagger \right].$$  \hspace{1cm} (B3)

We want to demonstrate that the following density matrix is an exact steady state:

$$\rho_{\infty} = \sum_{N} \pi_{N} \mathcal{I}_{N},$$  \hspace{1cm} (B4)

with

$$\pi_{N} = A \left( \frac{\Gamma_{\text{em}}}{\Gamma_{\text{loss}}} \right)^{N}.$$  \hspace{1cm} (B5)

First, since the hamiltonian preserves the total photon number, and that the density matrix is equal to identity of each sub Hilbert space with a defined photon number, we get that $[H, \rho_{\infty}] = 0$. Secondly for the Lindblad operators, the non hermitian hamiltonian terms have a simple action on the density matrix:

$$\rho_{\infty} \sum_{i} a_i^\dagger a_i = \rho_{\infty} \hat{N} = \hat{N} \rho_{\infty} = \sum_{i} a_i^\dagger a_i \rho_{\infty},$$  \hspace{1cm} (B6)

$$\rho_{\infty} \sum_{i} a_i^\dagger a_i = \rho_{\infty} \hat{N} = \hat{N} \rho_{\infty} = \sum_{i} a_i^\dagger a_i \rho_{\infty},$$  \hspace{1cm} (B7)

where $k$ is the number of cavities. Remain the special terms of the form $a_i^\dagger \rho a_i$:

$$\sum_{i} a_i^\dagger \rho_{\infty} a_i = \sum_{i} \sum_{N} \sum_{f(N)} \sum_{f(N) \neq f(N-1)} |f\rangle \langle f|.$$  \hspace{1cm} (B8)

In the same way, we find:

$$\sum_{i} \sum_{N} \sum_{f(N)} \sum_{f(N) \neq f(N-1)} |f\rangle \langle f|.$$  \hspace{1cm} (B9)

If we sum all contributions together, we get then a total zero contribution:

$$\mathcal{L}_{\text{loss}}(\rho_{\infty}) + \mathcal{L}_{\text{em}}(\rho_{\infty}) = \sum_{N} \sum_{f(N)} \pi_{N} \mathcal{I}_{N} + \pi_{N} \mathcal{I}_{N} \mathcal{I}_{N-1} + \pi_{N} \mathcal{I}_{N} \mathcal{I}_{N-2} + \cdots + \pi_{N} \mathcal{I}_{N} \mathcal{I}_{N-k} = 0.$$  \hspace{1cm} (B10)

**Appendix C: Corrections for coherences in the weakly non Markovian regime**

Here we demonstrate that the lowest-order correction with respect to the Grand canonical distribution in the weakly non Markovian regime presented in the section to the coherences between eigenstates (null in grand canonical statistics) are quadratic in the inverse pumping rate $\Gamma_{\text{pump}}^{-1}$ and not linear as a naive pertubative expansion would suggest. For this we calculate the first order contributions to the coherences of the operator $\delta \mathcal{M}$ (defined in eqs. A14, A14) applied to the grand canonic density matrix and show them to be 0. Let us
calculate first the contribution of the first two terms:

\[
\sum_i \langle f | \delta a_i^\dagger \rho_\infty a_i | f' \rangle = \sum_{i, f, f'} \langle f | \delta a_i^\dagger | \tilde{f} \rangle \langle \tilde{f} | a_i | f' \rangle 
\times \langle \tilde{f} | \rho_\infty | f' \rangle 
= \sum_{i, f} \langle f | \delta a_i^\dagger | f \rangle \langle f | a_i | f' \rangle \langle \tilde{f} | \rho_\infty | \tilde{f} \rangle.
\]

(C1)

In the same way:

\[
\sum_i \langle f | a_i^\dagger \rho_\infty \delta a_i | f' \rangle = \sum_{i, f} \langle f | a_i^\dagger | f \rangle \langle f | \delta a_i | f' \rangle \langle \tilde{f} | \rho_\infty | \tilde{f} \rangle
\]

(C2)

Then we know that

\[
\langle f | \delta a_i^\dagger | \tilde{f} \rangle = -\frac{i (\omega_f - \omega_{a_i})}{\Gamma_{\text{pump}}} \langle f | a_i^\dagger | \tilde{f} \rangle + \mathcal{O} \left( \frac{1}{\Gamma_{\text{pump}}} \right)^2
\]

(C3)

Let us choose a reference state \( | f_0 \rangle \) with the same photon number as \( | \tilde{f} \rangle \). Then \( \langle \tilde{f} | \rho_\infty | \tilde{f} \rangle = \langle f_0 | \rho_\infty | f_0 \rangle + \mathcal{O} \left( \frac{1}{\Gamma_{\text{pump}}} \right) \).

All these additional terms give second order contributions, and we do not consider them. Thus to the first order:

\[
\sum_i \langle f | \delta a_i^\dagger \rho_\infty a_i + a_i^\dagger \rho_\infty \delta a_i | f' \rangle
= \sum_{i, f} \langle f | a_i^\dagger | f \rangle \langle f | a_i | f' \rangle \langle \tilde{f} | \rho_\infty | \tilde{f} \rangle
\times \langle \tilde{f} | p_{\text{pump}} | f' \rangle
= \sum_{i, f} \langle f | a_i^\dagger a_i | f' \rangle
= \sum_{i, f} \langle f | a_i^{\dagger} a_i | f' \rangle
= \sum_{i, f} \langle f | N | f' \rangle
= N_f \delta f_f
= 0.
\]

(C4)

A similar reasoning allows to show that

\[
\sum_i \langle f | a_i \delta a_i^\dagger \rho_\infty + \rho_\infty \delta a_i a_i^\dagger | f' \rangle = 0.
\]

(C5)

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