

Variational Renormalization Group for Dissipative Spin-Cavity Systems: Periodic Pulses of Nonclassical Photons from Mesoscopic Spin Ensembles

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Mesoscopic spin ensembles coupled to a cavity offer the exciting prospect of observing complex nonclassical phenomena that pool the microscopic features from a few spins with those of macroscopic spin ensembles. Here, we demonstrate how the collective interactions in an ensemble of as many as a hundred spins can be harnessed to obtain a periodic pulse train of nonclassical light. To unravel the full quantum dynamics and photon statistics, we develop a time-adaptive variational renormalization group method that accurately captures the underlying Lindbladian dynamics of the mesoscopic spin-cavity system.

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Introduction.—In the past decade, there has been considerable interest in the development of hybrid quantum systems [1,2], where the interactions between spins or emitters with the modes of an electromagnetic field offer a cumulative advantage in designing quantum protocols, ranging from quantum many-body simulations [3,4] to the processing and storage of quantum information [5–8]. The majority of theoretical and experimental studies on such hybrid systems have focused on two very distinct regimes. On the one hand, macroscopic spin ensembles (SEs) and their collective properties have been investigated in the context of superradiance [9,10], amplitude bistability [11,12], spectral engineering [13,14], quantum memories [15–17], and suppression of decoherence through the cavity protection effect [18–20]. In this macroscopic limit, however, the light-matter interaction can be treated already on a semiclassical level [21], with possible quantum corrections [22,23]. On the other hand, in the microscopic limit, where a single or just a few spins couple to a cavity, this interaction demands full quantum solutions due to the anharmonicities of the excitations [24], resulting in exotic nonclassical phenomena such as antibunching [25], photon blockade [26], and single-photon emission [27]. Here we will explore the largely uncharted mesoscopic regime that offers the unique possibility to synergistically combine collective with nonclassical features that are otherwise restricted to the two separate regimes mentioned above. First signatures in this direction are already starting to emerge, such as through the observation of unconventional photon blockade [28–32], superbunching [33], and nonclassical photon bundles [34].

While experimental implementations of mesoscopic SEs are already within reach, especially using superconducting qubits [35], quantum dots [36], NV centers [37], rare-earth ensembles [38], and atomic gases [39,40], theoretical studies for such systems have been restricted to very specific

regimes, as the exponential growth of the Hilbert space limits any complete solution beyond a few spins. Most commonly, one is limited to either very weak excitations [41–43], few spin systems [44,45], or to ensembles without any inhomogeneous broadening [46–51]. Although these limits have already provided valuable insights into mesoscopic systems, they represent only the tip of the iceberg. There is definitely more to explore when going beyond these restrictions by taking into account the complex interplay between quantum effects, inhomogeneity, and nonlinearity due to excitations.

In this Letter, we formulate a powerful approach to investigate the full quantum dynamics of an inhomogeneous mesoscopic ensemble of as many as a hundred spins inside a quantum cavity, driven by a short coherent field. The spins are arranged such that their transition frequencies form a spectral frequency comb [52–54]. We demonstrate that the temporal evolution of such a comb-shaped ensemble results in a periodic and long-lived pulse train of nonclassical photons in the cavity. Here, the mesoscopic limit allows us to profit from an enhanced collective spin-cavity coupling, while also creating sub-Poissonian light fields due to the anharmonic nature of the excitations. In particular, the synergy of anharmonic and collective properties gives rise to periodic photon pulses operating close to the single-photon regime, which provides a valuable resource for quantum protocols such as linear optical quantum computing [55], single-photon cryptography [56], and low-light imaging [57]. In intervals between two photon pulses, the field also exhibits the exotic phenomenon of superbunching, which is often associated with correlations in the spin ensemble [33] or in the gain medium of quantum-dot microlasers [58]. Moreover, the strong driving in this regime also provides the exciting prospect of creating relatively high cavity photon numbers with nonclassical statistics. In turn,

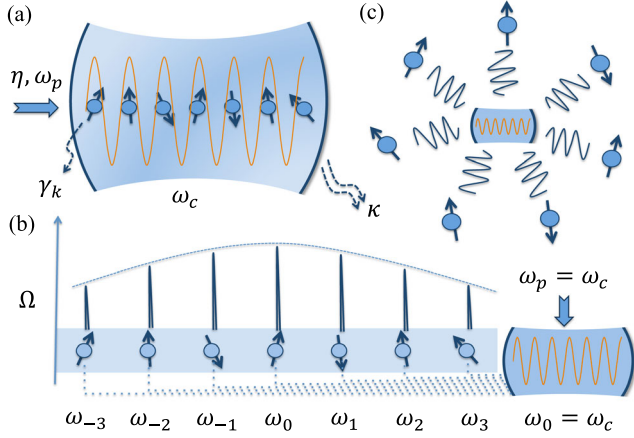


FIG. 1. (a) A mesoscopic spin ensemble interacting with a cavity, resonant at frequency ω_c . The cavity is driven by a coherent pulse of strength η and frequency ω_p . The cavity and spin losses are given by κ and γ_k . (b) The spin transition frequencies ω_j in the ensemble form a spectral frequency comb, with collective coupling strength Ω . (c) The spin-cavity system can be considered as a central body system.

however, the corresponding spin-cavity dynamics cannot be analyzed using known theoretical approaches. We thus develop a time-adaptive variational renormalization group method [59–61] that efficiently describes the Lindbladian dynamics of the mesoscopic spin-cavity system. For a coherent reading, we begin with a description of our model and the resulting physics before presenting our method.

Model.—An ensemble of N two-level emitters or spins inside a cavity, see Fig. 1(a), can be modeled using the Tavis-Cummings Hamiltonian [62], which under the dipole and rotating wave approximations reads

$$\mathcal{H} = \frac{1}{2} \sum_{k=1}^N \omega'_k \sigma_k^z + \omega_c \hat{a}_c^\dagger \hat{a}_c + i \sum_{k=1}^N g_k (\sigma_k^+ \hat{a}_c - \sigma_k^- \hat{a}_c^\dagger) + i[\eta(t) \hat{a}_c^\dagger e^{-i\omega_p t} - \eta^*(t) \hat{a}_c e^{i\omega_p t}], \quad (1)$$

where we take $\hbar = 1$. Here, ω_c is the resonance frequency of the cavity field \hat{a}_c , and ω'_k, g_k are the transition frequency and coupling strength for the k th spin (a spatial dependence of the spins can be included, but is not considered here). Furthermore, $\sigma_k^z, \sigma_k^+,$ and σ_k^- are the spin-1/2 Pauli operators. The quantum cavity is coherently driven with frequency ω_p and intensity $\eta(t)$. In general, the cavity and the spins in the mesoscopic ensemble are lossy, and the open dynamics is governed by the Lindblad equation, $d\rho/dt = \mathcal{L}[\rho] = -i[\mathcal{H}, \rho] + \kappa \mathcal{L}_{\hat{a}_c}[\rho] + \sum_k \gamma_k \mathcal{L}_{\sigma_k^-}[\rho]$, where $\mathcal{L}_{\hat{x}}[\rho] = \hat{x}\rho\hat{x}^\dagger - \frac{1}{2}\{\hat{x}^\dagger\hat{x}, \rho\}$ for $\hat{x} = \hat{a}_c$ and σ_k^- . The radiative losses of the cavity and spins are given by κ and γ_k . For macroscopic SEs, the spins and the cavity can both be treated semiclassically, and the expectation values are solved using the Maxwell-Bloch equations [21], and quantum corrections thereof [22,23]. However, to capture all the complex features of the quantum dynamics in

mesoscopic systems, the Lindblad equation needs to be solved exactly.

Nonclassical light in mesoscopic ensembles.—To demonstrate the complex nonclassical phenomena associated with mesoscopic spin-cavity interactions, we consider ensembles with up to $N = 105$ spins arranged in a finite spectral comb, with transition frequencies spaced at equidistant intervals. Such frequency combs have already been engineered in macroscopic ensembles, where the collective interactions result in long coherence times suitable for efficient quantum memory protocols [52,53] and long-lived pulses of classical light [54]. We demonstrate now explicitly that moving to a mesoscopic SE allows us to harness the quantum effects of light-matter interactions, thus making the pulses emitted from the mesoscopic SE nonclassical. Specifically, we propose a protocol for creating a periodic pulse train of antibunched light with sub-Poissonian photon statistics. The transition frequencies (ω'_k) of the spins in the spectral comb are arranged around the cavity frequency ω_c , with m (odd) distinct frequencies given by $\omega_j = \omega_c + j\Delta\omega$ for $j = \{-(m-1)/2, \dots, (m-1)/2\}$, as shown in Fig. 1(b). For an N -spin ensemble inside the cavity, each frequency ω_j in the spectral comb corresponds to a subensemble of $N' = N/m$ spins. The coupling constants for each of the subensembles and the cavity follow a Gaussian distribution, $\Omega_j = \Omega_0 \exp[-(\omega_c - \omega_j)^2/2\lambda^2]$, where Ω_0 is the coupling strength for the central subensemble, which is resonant with the cavity, and λ is the standard deviation of the distribution. Assuming that within each of the altogether $m = 7$ subensembles all the spins have the same coupling strength, i.e., $\Omega_j^2 = \sum_k^N g_{j,k}^2 = N' g_j^2$, the collective coupling of the total spin ensemble is given by $\Omega^2 = \sum_k \Omega_k^2$. We drive this hybrid quantum system resonantly with a short coherent pulse of intensity, $\eta(t) = \eta \in \text{Re}$, for $0 \leq t \leq t'$, and $\eta(t) = 0$, otherwise. Before the pulse arrives, the initial spin-cavity system is unexcited and the cavity is in the vacuum state. The coupling strengths and characteristic width of the comb are chosen as $\Omega_0/2\pi = 30$ MHz, $\lambda/2\pi = 150$ MHz, and $\Delta\omega/2\pi = 40$ MHz. The cavity and spin loss terms are taken as $\kappa/2\pi = 0.4$ MHz and $\gamma = \kappa/40$, respectively, with $\eta = 40\kappa$. The driving pulse duration t' is 1/5 of the characteristic timescale $2\pi/\Delta\omega$.

The first important feature of our mesoscopic frequency comb is the periodic pulse train of light it emits, exhibited by sharp revivals of the average cavity photon number $\langle \hat{a}_c^\dagger \hat{a}_c \rangle$, as shown in Fig. 2. Here, the peaks correspond to the collective transfer of excitations from the spin ensemble to the cavity, as evident from the sharp decrease in the spin excitation $\langle \sigma_i^+ \sigma_i^- \rangle_{\omega_i = \omega_c}$ at the revivals. The periodic pulses result from the constructive rephasing of spins in different subensembles of the comb, with the time interval between subsequent peaks commensurate with the inverse of the spectral width, $\Delta\tau \approx 2.2\pi/\Delta\omega = 173$ ns. This is a

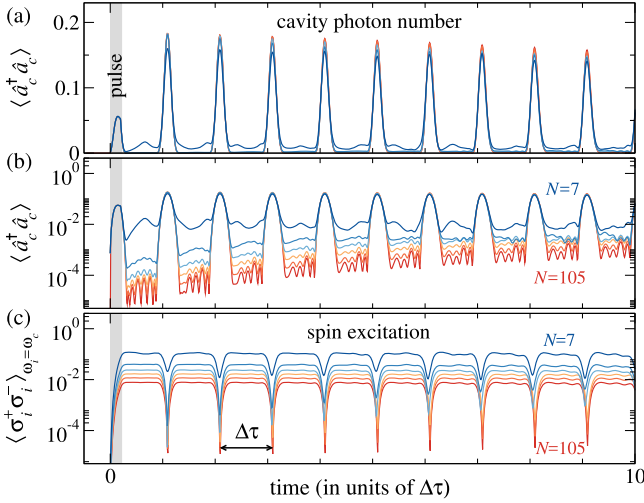


FIG. 2. Temporal evolution of the mesoscopic spin-cavity system. The figures show the cavity photon number $\langle \hat{a}_c^\dagger \hat{a}_c \rangle$ in (a) linear and (b) log scale, and (c) the spin excitation $\langle \sigma_i^+ \sigma_i^- \rangle_{\omega_i=\omega_c}$, varying with time, and for ensembles containing $N = 7, 21, 35, 49, 70$, and 105 spins, shown with colors varying from blue to red. The shaded region at times $0 \leq t \leq t'$ indicates the short rectangular driving pulse and $\Delta\tau$ is the interval between the periodic revivals.

hallmark of the collective behavior of the spins in the spectral frequency comb [63]. We note that during the transfer of energy from the cavity to the ensemble, larger ensembles, i.e., larger N , not only lead to enhanced coupling but also produce more stable and sharper photon pulses as excitations are distributed over more spins. In contrast, for few spins, significant photon excitations may also exist between the peaks, as observed in Fig. 2(b).

The second important, and in fact, central feature of these periodic light pulses is their distinct nonclassical character as inherent in their photon statistics. Using the equal-time second-order correlation function at time t , defined as $g_2(t) = \langle \hat{a}_c^{\dagger 2}(t) \hat{a}_c^2(t) \rangle / \langle \hat{a}_c^\dagger(t) \hat{a}_c(t) \rangle^2$, we observe in Fig. 3 that at times where the photon pulse arrives, the field is distinctly sub-Poissonian, i.e., $g_2(t) < 1$. While for all classical sources $g_2(t) \geq 1$ (unity for coherent light), sub-Poissonian light with $g_2(t) < 1$ is explicitly nonclassical. Here, we observe that such nonclassicality of the pulse train persists even for ensembles containing more than a hundred spins, as evidenced by $g_2^{\min} < 1$ in Fig. 3(c). Therefore, a relatively large mesoscopic ensemble can be used to generate a high-quality, nonclassical pulse train of photons. We note that there are interesting demarcations in the nonclassical nature of the photon pulse. To be specific, while the pulses are sub-Poissonian and distinctly nonclassical at all times (for all N), the unambiguous single-photon regime, $g_2^{\min} \ll 1$, is achieved only after a finite evolution time (which is shorter for smaller N).

We also explicitly checked that higher-order correlation functions, up to order $n = 4$, are also less than unity; i.e.,

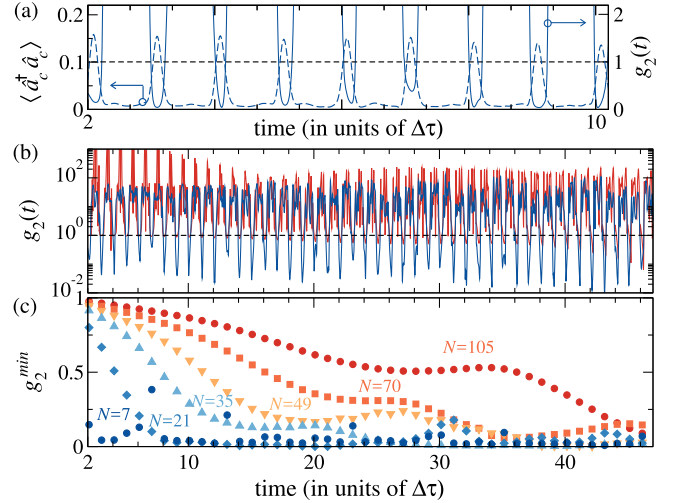


FIG. 3. Photon statistics of the transient cavity field (all parameters and color codings are the same as in Fig. 2). (a) Equal-time second-order correlation function $g_2(t)$ in comparison with the cavity photon number $\langle \hat{a}_c^\dagger \hat{a}_c \rangle$, for $N = 7$. (b) Temporal evolution of $g_2(t)$ for $N = 7$ and 105 . (c) Minimum value of $g_2(t)$ close to a pulse revival g_2^{\min} , for $N = 7, 21, 35, 49, 70$, and 105 . The horizontal black dashed line in (a) and (b) corresponds to $g_2(t) = 1$ for coherent light.

$g_n(t) = \langle \hat{a}_c^{\dagger n}(t) \hat{a}_c^n(t) \rangle / \langle \hat{a}_c^\dagger(t) \hat{a}_c(t) \rangle^n < 1$. Interestingly, at $t > 45\Delta\tau$, very low values of $g_n(t)$ ($n = 2, 3, 4$) are attained even for $N = 105$ (see Fig. 1 of the Supplemental Material [64]). In an experimental setting, such a suppression of multiphoton detection is considered to be a distinctive characteristic of single-photon emitters [68], making our system an interesting candidate for the design of quantum protocols [55–57]. An important feature is the persistent periodicity $\Delta\tau$ of the nonclassical pulse, which can be modulated by tuning the peak spacing $\Delta\omega$ in the spectral frequency comb. We note that this periodicity is present even for larger ensembles where antibunching is weak. For hybrid quantum systems, such a periodicity may allow for a temporal synchronization of the nonclassical light during an experimental phase, which is crucial for quantum memory protocols [16]. Another interesting cooperative behavior we observe is superbunching of the cavity field in intervals between the peaks, where $g_2(t) \gg 1$. This phenomenon has previously been related to superradiance arising from spin correlations in the ensemble [33,69]. Here, the low photon number in the superbunched emission is characteristic of the superradiant excitation being collectively stored in the spins rather than in the cavity. Such effects have also been reported in bimodal quantum-dot microlasers, where superbunching is induced by correlations in the gain medium [58] or irregular mode switching [70].

Renormalization for the Lindbladian dynamics.—To arrive at the above results we take advantage of the fact

that several exotic phenomena in spin-ensemble-cavity systems arise from Lindbladian dynamics that does not necessarily generate large correlations between the spins. This allows us to apply a time-adaptive variational renormalization group method [59–61], which efficiently maps the transient dynamics to a highly reduced vector space [71–74]. To set up this approach, we first map the system to a higher-dimensional complex vector space, such that a $d \times d$ density matrix ρ is vectorized to a d^2 superket, $|\rho\rangle = \text{vec}(\rho)$. Also, a $d \times d$ operator \hat{O} , which acts on ρ , is given by the higher-dimensional $d^2 \times d^2$ superoperator $\hat{\mathcal{O}}$, which now acts on $|\rho\rangle: \rho \rightarrow |\rho\rangle$, $\hat{O}\rho \rightarrow (\hat{O} \otimes \mathbb{I})|\rho\rangle = \hat{\mathcal{O}}|\rho\rangle$, and $\rho\hat{O} \rightarrow (\mathbb{I} \otimes \hat{O}^T)|\rho\rangle = \hat{\mathcal{O}}'|\rho\rangle$. The Lindblad equation in such a superoperator space is then given by $d|\rho\rangle/dt = \tilde{\mathcal{L}}|\rho\rangle$, where

$$\tilde{\mathcal{L}} = -i(\mathcal{H} \otimes \mathbb{I} - \mathbb{I} \otimes \mathcal{H}^T) + \kappa \tilde{\mathcal{L}}'_{\hat{a}_c} + \sum_k \gamma_k \tilde{\mathcal{L}}'_{\sigma_k}, \quad (2)$$

with $\tilde{\mathcal{L}}'_{\hat{x}} = \hat{x} \otimes \hat{x}^* - \frac{1}{2} \hat{x}^\dagger \hat{x} \otimes \mathbb{I} - \frac{1}{2} \mathbb{I} \otimes \hat{x}^T \hat{x}^*$. We note that in contrast to low-dimensional quantum spin systems with short-range interactions, all spins in the mesoscopic SE interact only with the cavity. Therefore, we can map the renormalization of the hybrid spin-cavity system to a central body problem [75], as illustrated in Fig. 1(c), albeit in a higher-dimensional superoperator space. Here, the cavity acts as the central quantum object that couples to each of the spins in the ensemble, which behaves like a bath in the superoperator space. The terms in \mathcal{H} and $\tilde{\mathcal{L}}'_{\hat{x}}$ [see Eq. (2)] can thus be written as a sum of individual spin-cavity terms, such that $\tilde{\mathcal{L}} = \sum_k \tilde{\mathcal{L}}_k$. A description of the mapping to the superoperator space is provided in Sec. I A of the Supplemental Material [64].

The two key steps in implementing the variational renormalization group for the Lindbladian dynamics are (i) the variational search for a truncated superoperator space and (ii) a time-adaptive Lindbladian evolution of the spin-cavity system. The former is obtained using the Schmidt decomposition, $|\rho\rangle = \sum_{\tilde{k}=1}^{\mathcal{K}} \alpha_{\tilde{k}} |\tilde{k}_A\rangle |\tilde{k}_B\rangle$, where the system is divided into blocks, A and B , as done during a renormalization method [60]. Here, $\{\alpha_{\tilde{k}}\}$ are the Schmidt coefficients in descending order, and $|\tilde{k}_A\rangle$ and $|\tilde{k}_B\rangle$ are the eigenvectors of the reduced superoperators of $|\rho\rangle$. \mathcal{K} is bounded from above by $r = \min[d_{\mathcal{R}_A}, d_{\mathcal{R}_B}]$, and is a measure of the total bipartite correlations [76]. Importantly, for several open systems $\alpha_{\tilde{k}}$ decays rapidly with \tilde{k} [71]. Thus, by retaining only the \mathcal{D} highest values of $\alpha_{\tilde{k}}$, we can approximate $|\rho\rangle$ and renormalize it to a significantly reduced dimension, i.e., $|\tilde{\rho}\rangle = \sum_{\tilde{k}=1}^{\mathcal{D}} \alpha_{\tilde{k}} |\tilde{k}_A\rangle |\tilde{k}_B\rangle$, where $\mathcal{D} \ll r$. The accuracy of the renormalization depends on the choice of \mathcal{D} and is exact for weakly or uncorrelated systems. For very high correlations, large values of \mathcal{D} need to be considered and the method is less efficient. To implement the Lindbladian evolution, we consider the dynamics

governed by $d|\rho\rangle/dt = \sum_k \tilde{\mathcal{L}}_k |\rho\rangle$. The superoperator space of the system is numerically renormalized and truncated at each step in a time-adaptive manner, using the Suzuki-Trotter decomposition [77]. This approach is comparable to a time-evolving block decimation [78,79] or a time-dependent density matrix renormalization group [80,81]. A detailed description of our method, error analysis, and a benchmark against exact solutions for few spins is provided in Secs. I–IV of the Supplemental Material [64].

For mesoscopic SEs in a cavity, solutions for the quantum dynamics have so far been achieved only for a few limited cases, such as for very weak excitations, where only a couple of low-excitation states are considered [41]. Alternatively, $\mathcal{L}[\rho]$ can be approximated by an effective Hamiltonian [42,43] in the weak excitation regime where quantum jumps are neglected. In turn, quantum trajectories include jumps but are limited to few spins [44,45]. Other methods involve direct solutions of $\mathcal{L}[\rho]$, using permutation symmetry for ensembles of identical spins [50,51], cumulant expansions for weakly correlated homogeneous ensembles [48,49], or approximate semiclassical solutions [46,47]. For the renormalization method we develop, the evolution is decomposed and exactly solved at the level of individual spin-cavity terms. This allows us to work in an extended parameter regime, with far more spins, higher number of excitations, and with inhomogeneous ensembles, which are typically not accessible using one of the above methods. Moreover, being based on the seminal Lindblad equation, our approach is distinct from those tensor-network methods that study open dynamics by simulating the unitary evolution of the larger system-environment states [82–84]. In particular, our approach does not require any additional restrictions on the environment beyond the master equation formalism. Our method is thus a powerful tool to obtain the transient or steady states of mesoscopic spin-cavity systems.

Conclusion and outlook.—We demonstrate that mesoscopic ensembles of spins coupled to a quantum cavity provide an interesting new platform for studying and tailoring nonclassical light fields. Based on recent experimental progress [1,2], implementing the proposed comb-shaped ensemble should be readily possible and an attractive option for creating a pulsed quantum source of light. These results provide just a first glimpse into the complex quantum dynamics of mesoscopic spin-cavity systems now accessible with the numerical method we introduce here. Our approach is based on the key insight that variational renormalization group and tensor network methods that have recently been successfully applied to low-dimensional quantum many-body systems [59–61] can be adapted to efficiently treat open spin-cavity systems. We thereby bridge a gap in the theoretical understanding of mesoscopic spin ensembles, and open up new directions to investigate complex parameter regimes that have remained out of reach so far.

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