Photon blockade in weakly-driven cavity QED systems with many emitters

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Abstract
We use the scattering matrix formalism to analyze photon blockade in coherently-driven CQED systems with a weak drive. By approximating the weak coherent drive by an input single- and two-photon Fock state, we reduce the computational complexity of the transmission and the two-photon correlation function from exponential to polynomial in the number of emitters. This enables us to easily analyze cavity-based systems containing ~50 quantum emitters with modest computational resources. Using this approach we study the coherence statistics of polaritonic photon blockade while increasing the number of emitters for resonant and detuned multi-emitter CQED systems — we find that increasing the number of emitters worsens photon blockade in resonant systems, and improves it in detuned systems. We also analyze the impact of inhomogeneous broadening in the emitter frequencies on both polaritonic and subradiant photon blockade through this system.

Introduction. Cavity quantum electrodynamics (CQED) is a fundamental model of light and matter interaction whose experimental implementations allow for a high degree of control. Atomic and solid state CQED systems with a few two-level emitters have exhibited a rich set of quantum phenomena in transmission statistics, including, but not limited to, the vacuum Rabi oscillations [1, 2], the conventional and the unconventional photon blockade [3–5], and the photon-induced tunneling [6]. Extending this research to more complex systems is expected to unveil a new set of coherent effects [7, 8]. While suitable approximations can provide understanding of the eigenstructure of multi-element CQED systems [9, 10] obtained in experiments [11, 12], the numerical studies of light-emission and scattering from this system have been limited due to the exponential scaling of the Hilbert space with the number of emitters.

The scattering matrix provides the solution to this problem. It is a fundamental object representing the response of a quantum system described by a field theory. Recently, a general formalism for computing this scattering matrix for an arbitrary time-independent and time-dependent Markovian quantum-optical system was developed [13, 14]. In particular, the scattering matrix was expressed in terms of the Green’s functions of the quantum-optical system, which can be computed entirely within the Hilbert space of the quantum-optical system. Use of the scattering matrices allows relating the transmission and two-photon correlation through a system to the single- and two-photon scattering matrix — computation of which can be performed in polynomial time \( \sim O(N^5) \) in the number of emitters \( N \).

In this Letter, we use the scattering matrix formalism to study multi-emitter CQED systems with a large number of emitters (\( N \sim 50 \)) driven by weak continuous-wave classical light (e.g. a laser). We numerically study the multi-emitter CQED system to analyze the impact of increasing the number of emitters on its scattering properties. In contrast to the \( \sqrt{N} \)-fold increase in the coupling strength, we show that increasing the number of emitters does not increase the depth of the photon blockade in a resonant multi-emitter CQED systems with identical emitters. However, we find that multi-emitter-cavity interaction does provide an additional phase shift to increase the depth of photon blockade if the emitters are detuned from the cavity resonance. Finally, we consider an experimentally realistic setting wherein the emitter frequencies are inhomogeneously broadened (e.g. if the system is implemented as a diamond [15–17] or silicon carbide [18–21] nanocavity incorporating color centers) and analyze polaritonic and subradiant photon blockade in both resonant and detuned multi-emitter systems.

Simulation Method. A schematic of the considered system is shown in Fig. 1 — a cavity, with annihilation operator \( a \), is coupled to \( N \) two-level emitters, with lowering operators \( \sigma_i \), \( 1 \leq i \leq N \). The cavity is excited through a waveguide, with a frequency dependent annihilation operator \( b_\omega \), and the emission from the cavity is collected through another waveguide, with annihilation operator \( c_\omega \). The emitters, in addition to coupling to the cavity mode, also radiate into loss channels with annihilation operators \( i^{(s)}_\omega \) — these loss channels model the linewidths of the emitters. The Hamiltonian for the multi-emitter

\[ H = \sum_{\omega, i} \left( \epsilon_i \sigma_i^+ \sigma_i - \frac{1}{2} \left( b_\omega \sigma_i^+ + c_\omega \sigma_i \right) \right) + \sum_{\omega} \left( \gamma_\omega i^{(s)}_\omega c_\omega + \text{H.c.} \right) + \sum_{\omega, i} \kappa_{\omega i} \left( b_\omega \sigma_i + \text{H.c.} \right). \]
CQED system is given by:

\[ H_{\text{sys}} = \omega_c a^\dagger a + \sum_{i=1}^{N} \left( \omega_i \sigma_i^\dagger \sigma_i + g_i (a \sigma_i^\dagger + \sigma_i a^\dagger) \right). \]  

(1)

where \( \omega_c \) is the cavity resonance frequency, \( \omega_i \) is the transition frequency of the \( i \)-th emitter and \( g_i \) is the coupling constant between the \( i \)-th emitter and the cavity mode. We study the excitation of this system with a continuous-wave coherent state at frequency \( \omega_L \). In the framework of scattering theory, this corresponds to exciting the system with an input state \( |\psi_{\text{in}}\rangle \) given by:

\[ |\psi_{\text{in}}\rangle = \exp[\beta_0 (b_\omega L - b_{-\omega L})] |\text{vac}\rangle \]  

(2)

where \( \beta_0^2 \) is the photon flux (number of photons per unit time) in the input coherent state. As is detailed in the supplementary material, we establish the following relationship of the transmission \( T(\omega_L) \) and two-photon correlation \( g^{(2)}(t_1, t_2; \omega_L) \) to the single-photon \( S_{\text{c},c}(\cdot) \) and two-photon \( S_{\text{c},c}(\cdot) \) scattering matrices for continuous-wave input in the limit of small input photon flux \( \beta_0^2 \):

\[ T(\omega_L) = \left| \int_{t'=\infty}^{\infty} S_{\text{c}}(t; t') \exp(-i\omega_L t') dt' \right|^2 \]  

(3a)

\[ g^{(2)}(t_1, t_2; \omega_L) = \frac{1}{4 \Gamma_0^2(\omega_L)} \times \]  

\[ \left| \int_{t'_1, t'_2 = -\infty}^{\infty} S_{\text{c},c}(t_1, t_2; t'_1, t'_2) \exp(-i\omega_L (t'_1 + t'_2)) dt'_1 dt'_2 \right|^2 \]  

(3b)

where the \( S \) matrices capture scattering of photons propagating in the input-waveguide (with annihilation operator \( b_\omega \)) to the output-waveguide (with annihilation operator \( c_\omega \)). The scattering matrices are functions only of the system operators and external coupling constants \( \kappa_{\text{c}} \) and \( \gamma_n \).

The dominant cost for computing these scattering matrices is that of diagonalizing the effective Hamiltonian \( H_{\text{eff}} \) (refer to the accompanying supplementary material for details):

\[ H_{\text{eff}} = H_{\text{sys}} - \frac{i\kappa}{2} a^\dagger a - \sum_{n=1}^{N} \frac{i\gamma_n}{2} \sigma_n^\dagger \sigma_n \]  

(4)

where \( \kappa = \kappa_b + \kappa_c \) is the total decay rate for the optical cavity. Since \( H_{\text{eff}} \) conserves the total excitation number \( (a^\dagger a + \sum_{n=1}^{N} \sigma_n^\dagger \sigma_n) \), this diagonalization can be performed separately within the excitation conserving subspaces of the full Hilbert space. For example, when computing the single- and two-photon scattering matrices, it is only necessary to diagonalize the effective Hamiltonian within the single- and two-excitation subspaces. Then, the cost of computation approximately scales as \( \sim O(N^6) \), where \( N \) is the number of emitters.

Results. Using a large number of identical emitters coupling coherently to the same cavity mode is a potential strategy to achieve strong coupling between the emitters and the cavity in a situation where an individual emitter only weakly couples to the cavity mode. Figure 2 shows the transmissivity \( T(\omega_L) \) and equal-time correlation \( g^{(2)}(0; \omega_L) \) for multi-emitter CQED systems with 1–50 emitters. We observe that the splitting between the polaritonic peaks in the transmissivity increases with the number of emitters on resonance with the optical mode — in particular, it scales as \( \sqrt{N} \), where \( N \) is the number of emitters. This is consistent with the result obtained on a direct diagonalization of \( H_{\text{sys}} \) within the first excitation subspace, which predicts a splitting of \( 2g\sqrt{N} \) between the eigenvalues of polaritonic eigenstates. We also observe that the minimum two-photon correlation \( g^{(2)}(0; \omega_L) \), which is achieved at the polaritonic frequencies, trends towards unity with an increase in the number of emitters for both strongly-coupled emitters.
1. Right of the Fano dip at the emitter-like polariton. Here, the light antibunches \((g^{(2)}(0; \omega_L) < 1)\) owing to the standard polaritonic photon blockade of a detuned system \([22, 23]\). With increasing number of emitters, the blockade effect degrades just as with the resonant CQED system.

2. Exactly at the Fano dip. Here, the light bunches \((g^{(2)}(0; \omega_L) > 1)\), which occurs due to scattering almost entirely from the connected (nonlinear) part of the two-photon \(S\) matrix. Notably for the detuned system, this two-photon bound state is essentially that of a bare two-level system \([24]\).

3. Slightly left of the Fano dip. Here, the light antibunches again due to intensity interference between the two-photon bound state and the unconnected (linear) part of the \(S\) matrix. The blockade depth increases with increasing number of emitters because the multi-emitter transmission spectra decreases the q-factor of the Fano resonance. This causes the phase relation between the connected and unconnected portions of the \(S\) matrix to change. The changing phase relation further causes oscillations in the delayed intensity coherence \([Fig. 3(b)]\).

While the previous analysis was primarily done under the assumption of identical emitters, practical multi-emitter CQED systems almost always have emitters with slightly different resonant frequencies — this non-ideality is termed as inhomogeneous broadening in the emitter frequencies. For solid-state color centers (silicon vacancies in 4H-SiC or SiV centers in diamond) embedded in a nanocavity, the distribution of the emitter frequencies can be modelled as a normal distribution with standard deviation \(\Delta \lesssim 20\) GHz \([18, 20, 21]\). Results of a Monte–Carlo analysis on the transmission and equal-time two-photon correlation through the multi-emitter system are shown in \(Fig. 4(a)\) for resonant emitters and \(Fig. 4(b)\) for detuned emitters, with these emitters each being weakly coupled to the cavity. We observe an emergence of a large number of very narrow linewidth dips in \(g^{(2)}(0; \omega_L)\) which correspond to the subradiant photon blockade that has been studied in CQED systems with two non-identical emitters \([8]\). The subradiant photon blockade dips reach very low \(g^{(2)}(0; \omega_L)\) values even for emitters that individually couple to the cavity only weakly. Moreover, for the resonant system, the distribution of the frequencies of the blockade dips \((\omega_B)\) reveal that the spread in the frequencies of the subradiant photon blockade is of the order of the inhomogeneous broadening in the emitter frequencies, whereas the frequencies of polaritonic photon blockade are significantly more robust to inhomogeneous broadening in the emitter frequencies albeit with a much larger value of \(g^{(2)}(0; \omega_B)\). A similar trend is observed in the detuned system \([Fig. 4(b)]\), with the polaritonic dip
FIG. 4. Impact of inhomogeneous broadening on the photon blockade in multi-emitter CQED systems for (a) the emitters, on an average, being resonant with the cavity. (b) emitters that are, on an average, detuned from the cavity resonance by \( \langle \omega_e \rangle - \omega_c = 0.8\kappa = 2\pi \cdot 20 \text{ GHz} \). For both cases, we show a typical lineshape \( g^{(2)}(0; \omega_L) \), and the statistics of the frequencies \( \omega_B \) and the \( g^{(2)}(0; \omega_B) \) values for the polaritonic and subradiant photon blockade (Note that the y-axis in the histogram is the unnormalized frequency of occurrence of the sample statistic). Parameter values \( \Delta = 25 \text{ GHz}, \kappa = 2\pi \cdot 25 \text{ GHz}, g = 0.2\kappa = 2\pi \cdot 5 \text{ GHz} \) and \( \gamma = 2\pi \cdot 0.3 \text{ GHz} \) are assumed in all simulations.

being much more sensitive to the inhomogeneous broadening in the emitter frequencies due the polariton being emitter-like in nature, while reaching very low \( g^{(2)}(0; \omega_B) \) values (\( \sim 0 - 0.1 \)) similar to the identical-emitter system. Our simulations seem to indicate the presence of a fundamental trade-off between the extent of the photon-blockade (i.e. how small is \( g^{(2)}(0; \omega_B) \)) and the robustness of the blockade frequency \( \omega_B \) to inhomogeneous broadening in the emitters. This analysis will be important for experimentalists interested in observing the photon statistics of multi-emitter CQED systems.

In conclusion, we used the scattering matrix formalism to calculate the transmissivity and the two-photon correlation in a weakly-driven multi-emitter CQED system, reducing the computational complexity form exponential to polynomial with respect to the number of emitters. This approach allowed us to easily study photon blockade in multi-emitter systems with up to 50 emitters. We showed that increasing the number of identical emitters has an interesting and rich physics in the transmission statistics. Moreover, we analyzed the impact of inhomogeneous broadening typical of color center emitters on the light scattered by the multi-emitter system. Finally, we find that the inhomogeneous broadening does not mask away polaritonic photon blockade in systems with solid-state vacancy centers.

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Supplementary information: Photon Blockade in weakly-driven CQED systems with many emitters

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ANALYZING TRANSMISSION STATISTICS FOR ARBITRARY BOSONIC SCATTERING PROBLEMS

In this section, we show that for any scattering problem with coherent drive, the transmissivity and two-particle correlation through the system can be expressed in terms of the single- and two-particle scattering matrices. Henceforth, we will consider the particles as photons without loss of generality. Photonic transport through multi-emitter systems like the ones considered in the main text are a special case of this general problem.

The main issue addressed in this section is the fact that a continuous wave coherent state input is not normalizable, with the photon number in the coherent state being infinitely large even at weak driving amplitudes. Intuitively, analyzing the response of a system to such a state should require computation of scattering matrices with an arbitrary number of input photons. Here, we show that despite this issue of normalizability of the coherent state, such a few-photon approximation of the continuous wave coherent state will still give the correct result for the transmission and the two-photon correlation in the limit of a weak coherent drive.

We consider the input state $|\psi_{in}\rangle$ to be a pulsed-coherent state given by:

$$|\psi_{in}\rangle = \exp \left( -\frac{1}{2} \int_{-\infty}^{\infty} |\beta(t)|^2 dt \right) \left[ |\text{vac}\rangle + \sum_{k=1}^{\infty} \frac{1}{k!} \int_{-\infty}^{\infty} dt_1 ... dt_k \beta(t_i) b_i^\dagger |\text{vac}\rangle \right]$$

(1)

where $\beta(t) = \beta_0 \text{rect}(t/\tau) \exp(-i \omega_L t)$, with $\text{rect}(t) = 1$ if $|t| \leq 1$ and 0 otherwise. In the limit of $\tau \to \infty$, this state approaches the continuous wave coherent state at frequency $\omega_L$. The output state is then given by:

$$|\psi_{out}(\omega_L, \tau)\rangle = \exp(-|\beta_0|^2 \tau) \left[ |\text{vac}\rangle + \sum_{k=1}^{\infty} \frac{\beta_0^k}{k!} |\psi_k(\omega_L, \tau)\rangle \right]$$

(2)

where

$$|\psi_k(\omega_L, \tau)\rangle = \frac{\beta_0^{-k}}{k!} \sum_{\mu_1, \mu_2, ... \mu_k} \int_{-\infty}^{\infty} dt_1 ... dt_k -\int_{t_1'}^{t_2'} ... dt_k' = -\int_{t_1'}^{t_2'} ... dt_k' \sum_{i=1}^{k} S_{\mu_1, \mu_2, ... \mu_k}(t_1, t_2 ... t_k; t_1', t_2' ... t_k') \prod_{i=1}^{k} \beta(t_i^\dagger) \beta(t_i) dt_i dt_i'$$

(3)

where $\mu_i$ denotes a waveguide or loss channel (i.e. $\mu_i \in \{ b, c, l(1), l(2), ... l(k) \}$), and $S_{\mu_1, \mu_2, ... \mu_k}(t_1, t_2 ... t_k; t_1', t_2' ... t_k')$ is the scattering matrix element which captures the scattering of $k$ photons in the input waveguide to $k$ photons in the ports $\mu_1, \mu_2, ... \mu_k$. We note that in the limit of $\tau \to \infty$, the integral with respect to $t_1', t_2' ... t_k'$ in Eq. 3 converges. To see this, note that this integral can equivalently be expressed in terms of the frequency-domain scattering matrix:

$$\lim_{\tau \to \infty} \int_{t_1'}^{t_2'} ... dt_k' S_{\mu_1, \mu_2, ... \mu_k}(t_1, t_2 ... t_k; t_1', t_2' ... t_k') \exp[-i \omega_L (t_1' + t_2' + ... + t_k')] dt_1' dt_2' ... dt_k'$$

$$= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S_{\mu_1, \mu_2, ... \mu_k}(\omega_1, \omega_2 ... \omega_k; \omega_L, \omega_L ... \omega_L) \exp[-i(\omega_1 t_1 + \omega_2 t_2 + ... + \omega_k t_k)] d\omega_1 d\omega_2 ... d\omega_k$$

(4)

As is outlined in [1], the general structure of the $k$-photon scattering matrix has at most $k$ delta-functions that conserve the total frequency of the input and output photons. Therefore, the $k$ integrals over the input-frequencies in Eq. 4 remove all the delta functions, resulting in a completely well-defined and finite integrand.

Finally, also note that for time-independent Hamiltonians such as the multi-emitter CQED system considered in the main text, the $k$ photon scattering matrix $S_{\mu_1, \mu_2, ... \mu_k}(t_1, t_2 ... t_k; t_1', t_2' ... t_k')$ depends only on the differences between the time-arguments, and not on their actual values i.e.

$$S_{\mu_1, \mu_2, ... \mu_k}(t_1, t_2 ... t_k; t_1', t_2' ... t_k') \equiv S_{\mu_1, \mu_2, ... \mu_k}(0, t_2 - t_1 ... t_k - t_1; t_1' - t_1, t_2' - t_1 ... t_k' - t_1)$$

(5)
Transmittivity with a weak continuous-wave coherent drive

Consider now the computation of the transmissivity $T(\omega; t, \tau)$ through the multi-emitter CQED system at amplitude $\beta_0$:

$$T(\omega_L; t, \tau, \beta_0) = \frac{\langle \psi_{\text{out}}(\omega_L, \tau) | c_1^\dagger c_2 | \psi_{\text{out}}(\omega_L, \tau) \rangle}{|\beta_0|^2} = \exp(-2|\beta_0|^2\omega) \sum_{k=1}^{\infty} \frac{|\beta_0|^{2k}}{(k!)^2} \langle \psi_k(\omega_L, \tau) | c_1^\dagger c_2 | \psi_k(\omega_L, \tau) \rangle$$  \hspace{1cm} (6)

wherein we have divided the photon flux (number of photons per unit time) in the input state in the input waveguide with the photon flux in the output state in the output waveguide at time $t$. Taking the limit of $\beta_0 \to 0$:

$$\lim_{\beta_0 \to 0} T(\omega_L; t, \tau, \beta_0) = \langle \psi_1(\omega_L, \tau) | c_1^\dagger c_2 | \psi_1(\omega_L, \tau) \rangle.$$  \hspace{1cm} (7)

Now, taking the limit of $\tau \to \infty$, we obtain:

$$\lim_{\tau \to \infty} \left[ \lim_{\beta_0 \to 0} T(\omega_L; t, \tau, \beta_0) \right] = \left| \int_{t_1'}^{-\infty} S_c(t_1; t_1') \exp(-i\omega_L t_1') dt_1' \right|^2.$$  \hspace{1cm} (8)

Finally, using the time-invariance of the system (Eq. 5), we immediately see that this limit is independent of $t$:

$$T(\omega_L) = \lim_{\tau \to \infty} \left[ \lim_{\beta_0 \to 0} T(\omega_L; t, \tau, \beta_0) \right] = \left| \int_{t_1'}^{-\infty} S_c(0; t_1', t_1) \exp(-i\omega_L t_1') dt_1' \right|^2 = \left| \int_{\tau'}^{-\infty} S_c(0; \tau') \exp(-i\omega_L \tau') d\tau' \right|^2.$$  \hspace{1cm} (9)

Two-photon correlation with a weak continuous-wave drive

The two-photon correlation in the output state is defined by:

$$g^{(2)}(t_1, t_2; \omega_L, \beta_0, \tau) = \frac{\langle \psi_{\text{out}}(\omega_L, \tau) | c_1^\dagger c_2 c_1 c_2 | \psi_{\text{out}}(\omega_L, \tau) \rangle}{\langle \psi_{\text{out}}(\omega_L, \tau) | c_1^\dagger c_1 | \psi_{\text{out}}(\omega_L, \tau) \rangle \langle \psi_{\text{out}}(\omega_L, \tau) | c_2^\dagger c_2 | \psi_{\text{out}}(\omega_L, \tau) \rangle} = \frac{\exp(2|\beta_0|^2\omega) \sum_{k=1}^{\infty} |\beta_0|^{2k} \langle \psi_k(\omega_L, \tau) | c_1^\dagger c_1 c_2 c_2 | \psi_k(\omega_L, \tau) \rangle}{\sum_{k=1}^{\infty} \langle \psi_k(\omega_L, \tau) | c_1^\dagger c_1 | \psi_k(\omega_L, \tau) \rangle \langle \psi_k(\omega_L, \tau) | c_2^\dagger c_2 | \psi_k(\omega_L, \tau) \rangle}.$$  \hspace{1cm} (10)

Taking the limit of $\beta_0 \to 0$ (corresponding to a weak coherent drive), we obtain:

$$\lim_{\beta_0 \to 0} g^{(2)}(t_1, t_2; \omega_L, \beta_0, \tau) = \frac{\langle \psi_2(\omega_L, \tau) | c_1^\dagger c_2 c_1 c_2 | \psi_2(\omega_L, \tau) \rangle}{4 \langle \psi_1(\omega_L, \tau) c_1^\dagger c_1 | \psi_1(\omega_L, \tau) \rangle \langle \psi_1(\omega_L, \tau) | c_2^\dagger c_2 | \psi_1(\omega_L, \tau) \rangle}.$$  \hspace{1cm} (11)

Next, we take the limit of $\tau \to \infty$ (corresponding to a continuous-wave drive) to obtain. As already shown in the previous subsection,

$$\lim_{\tau \to \infty} \langle \psi_1(\omega_L, \tau) | c_1^\dagger c_2 | \psi_1(\omega_L, \tau) \rangle = \left| \int_{\tau'}^{-\infty} S_c(0; \tau') \exp(-i\omega_L \tau') d\tau' \right|^2 = T(\omega_L).$$  \hspace{1cm} (12)

Similarly,

$$\lim_{\tau \to \infty} \langle \psi_2(\omega_L, \tau) | c_1^\dagger c_2 c_1 c_2 | \psi_2(\omega_L, \tau) \rangle = \left| \int_{t_1'}^{-\infty} \int_{t_1''}^{-\infty} S_{c,c}(t_1, t_2; t_1', t_1'') \exp[-i\omega_L (t_1' + t_1'')] dt_1' dt_1'' \right|^2 = \left| \int_{\tau_1', \tau_1''}^{-\infty} S_{c,c}(0, t_2 - t_1; \tau_1', \tau_1'') \exp[-i\omega_L (\tau_1' + \tau_1'')] d\tau_1' d\tau_1'' \right|^2.$$  \hspace{1cm} (13)
wherein we have used the time-invariance of the multi-emitter system in the last step. Therefore, the two-photon correlation, in the continuous-wave limit, depends only on the difference between the time-instants at which it is being computed. The complete expression for the two-photon correlation is given below:

\[
g^{(2)}(t_1, t_2; \omega_L) = \lim_{\tau \to \infty} \lim_{\beta_0 \to 0} g^{(2)}(t_1, t_2; \omega_L, \beta_0, \tau)
\]

\[
= \frac{1}{4T^2(\omega_L)} \left| \int_{t_1, t_2 = -\infty}^{\infty} S_{ee}(t_1, t_2; t_1', t_2') \exp[-i\omega_L(t_1' + t_2')] \, dt_1' \, dt_2' \right|^2
\]

\[
= \frac{1}{4T^2(\omega_L)} \left| \int_{\tau_1, \tau_2 = -\infty}^{\infty} S_{ee}(0, t_2 - t_1; \tau_1', \tau_2') \exp[-i\omega_L(\tau_1' + \tau_2')] \, d\tau_1' \, d\tau_2' \right|^2.
\]

(14a, 14b)

We briefly note that this limit can be taken for computing any arbitrary \( g^{(n)} \) in a similar way.

STRUCTURE OF THE SINGLE- AND TWO-PHOTON SCATTERING MATRICES

In this appendix, we outline the computation of the single- and two-photon scattering matrices \( (S_c(t_1; t_1') \) and \( S_{ee}(t_1, t_2; t_1', t_2') \) which are required for computing the transmission and two-photon correlations through the multi-emitter CQED system discussed in the main text. Specifically, we show that cost of computing these scattering matrices is dominated by the cost of diagonalizing the effective Hamiltonian, \( H_{eff} \), given by:

\[
H_{eff} = \left( \omega_c - \frac{i\kappa}{2} \right) a^\dagger a + \sum_{n=1}^N \left( \omega_n - \frac{i\gamma_n}{2} \right) \sigma_n^\dagger \sigma_n + \sum_{n=1}^N g_n (a \sigma_n^\dagger + \sigma_n a^\dagger),
\]

within the single- and two-excitation subspaces of the multi-emitter CQED system.

As is shown in [1, 2], these scattering matrices can be computed by computing the expectations of the cavity annihilation and creation operator \( (a \) and \( a^\dagger \)) evolved under the effective Hamiltonian \( H_{eff} \) of the multi-emitter system (Eq. 15):

\[
S_c(t_1; t_1') = -\sqrt{\kappa_b \kappa_c} \langle \phi | T \{ \hat{a}(t_1) \hat{a}^\dagger(t_1') \} | \phi \rangle
\]

\[
S_{ee}(t_1, t_2; t_1', t_2') = \kappa_b \kappa_c \langle \phi | T \{ \hat{a}(t_1) \hat{a}(t_2) \hat{a}^\dagger(t_1') \hat{a}^\dagger(t_2') \} | \phi \rangle
\]

where \( | \phi \rangle = |0, g_1, g_2 \ldots g_N \rangle \) is the ground state of the multi-emitter system, \( T \) indicates chronological ordering which, for a given set of time indices, orders the operators in decreasing order of the time indices and \( \hat{a}(t) \) and \( \hat{a}^\dagger(t) \) are given by:

\[
\begin{bmatrix}
  a(t) \\
  a^\dagger(t)
\end{bmatrix} = \exp(iH_{eff}t) \begin{bmatrix}
  a \\
  a^\dagger
\end{bmatrix} \exp(-iH_{eff}t).
\]

(16a, 16b, 17)

To proceed further, we note that the effective hamiltonian \( H_{eff} \) commutes with and hence conserves the total excitation number operator \( n \):

\[
n = a^\dagger a + \sum_{i=1}^N \sigma_i^\dagger \sigma_i.
\]

(18)

Therefore, the effective Hamiltonian and the number operator can be simultaneously diagonalized — in particular, the effective Hamiltonian \( H_{eff} \) can be expressed as a matrix:

\[
H_{eff} ≡ \begin{bmatrix}
  0 & 0 & 0 & 0 & \ldots \\
  0 & \text{diag}(\lambda_1) & 0 & 0 & \ldots \\
  0 & 0 & \text{diag}(\lambda_2) & 0 & \ldots \\
  0 & 0 & 0 & \text{diag}(\lambda_3) & \ldots \\
  \vdots & \vdots & \vdots & \vdots & \ddots
\end{bmatrix}
\]

(19)

where \( \lambda_i \) is a vector of eigenvalues of \( H_{eff} \) corresponding to its eigen-vectors within the \( i \)th excitation subspace. It can also be noted that the dimension of the \( i \)th subspace is \( O(N^i) \), where \( N \) is the number of emitters in the multi-emitter system. The operators \( a \) and \( a^\dagger \) can also be expressed as a matrix on the basis that diagonalizes \( H_{eff} \) and \( n \).
simultaneously. Since $a\ (a^\dagger)$ reduces (increases) the number of photons inside the cavity, and hence the number of
excitations, by 1, its matrix representation has the form:

$$a \equiv \begin{bmatrix} 0 & A_{0,1} & 0 & 0 & \cdots \\ 0 & 0 & A_{1,2} & 0 & \cdots \\ 0 & 0 & 0 & A_{2,3} & \cdots \\ \vdots & \vdots & \vdots & \vdots & \ddots \end{bmatrix} \quad \text{and} \quad a^\dagger \equiv \begin{bmatrix} 0 & 0 & 0 & \cdots \\ A_{0,1}^\dagger & 0 & 0 & \cdots \\ 0 & A_{1,2}^\dagger & 0 & \cdots \\ 0 & 0 & A_{2,3}^\dagger & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix}$$

(20)

where $A_{i,i+1}$ is the projection of $a$ onto the direct sum of the $i^{th}$ and $(i+1)^{th}$ excitation subspace expressed on the
basis that diagonalizes the effective Hamiltonian. Once we have computed this representation of $H_{\text{eff}}, a$ and $a^\dagger$, it is
straightforward to compute the scattering matrices and hence the transmission and two-photon correlation induced by the multi-emitter CQED system.

**Computation of the single photon scattering matrix and transmission**

Noting that $\exp(-iH_{\text{eff}}t)\ket{\phi} = \ket{\phi}$, the single-photon scattering matrix (Eq. 16a) can be expressed as:

$$S_{c}(t_1; t_1') = -\sqrt{\kappa_0\kappa_c} \langle \phi | a \exp(-iH_{\text{eff}}(t_1 - t_1')a^\dagger \rangle \theta(t_1 - t_1')$$

$$= -\sqrt{\kappa_0\kappa_c} A_{0,1} D \{ \exp(-i\lambda_1(t_1 - t_1')) \} A_{0,1}^\dagger \theta(t_1 - t_1')$$

(21)

where $D\{ \cdot \}$ of a vector $v$ is a diagonal matrix with elements of $v$ on its diagonal. Using Eq. 9, we obtain the following expression for the transmittivity:

$$T(\omega_L) = \kappa_0\kappa_c \left| \begin{array}{c} 1 \\ \lambda_1 - \omega_L \end{array} \right| A_{0,1}^\dagger \right|^2.$$

(22)

We note that this expression for transmission has an intuitively expected form — the $\omega_L - \lambda_1$ term introduces resonances at the frequencies that match the eigen-values $\lambda_1$, with the strength of the resonances depending on the matrix element of the cavity annihilation operator.

**Computation of the two-photon scattering matrix and two-photon correlation**

Note from Eq. 16b that the two-photon scattering matrix is symmetric with respect to an exchange of the time indices $t_1$ and $t_2$, and $t_1'$ and $t_2'$. Therefore, without loss of generality, we will assume $t_1 \geq t_2$ and $t_1' \geq t_2'$. The two-photon scattering matrix then reduces to:

$$S_{c,c}(t_1, t_2; t_1', t_2')$$

$$= -\kappa_0\kappa_c \left\{ \begin{array}{cc} \langle \phi | a \exp(-iH_{\text{eff}}(t_1 - t_2))a \exp(-iH_{\text{eff}}(t_2 - t_1'))a^\dagger \exp(-iH_{\text{eff}}(t_2' - t_1'))a^\dagger \rangle \theta(t_1 - t_2) \theta(t_1' - t_2') & \text{if } t_1 \geq t_2 \geq t_1' \geq t_2' \\ \langle \phi | a \exp(-iH_{\text{eff}}(t_1 - t_2))a \exp(-iH_{\text{eff}}(t_2 - t_1'))a \exp(-iH_{\text{eff}}(t_2' - t_1'))a^\dagger \rangle \theta(t_1 - t_2) \theta(t_1' - t_2') & \text{if } t_1 \geq t_1' \geq t_2 \geq t_2' \\ 0 & \text{otherwise} \end{array} \right\}$$

$$= -\kappa_0\kappa_c \left\{ \begin{array}{cc} A_{0,1} D \{ \exp(-i\lambda_1(t_1 - t_2)) \} A_{1,2}^\dagger D \{ \exp(-i\lambda_1(t_2' - t_1')) \} A_{0,1}^\dagger & \text{if } t_1 \geq t_2 \geq t_1' \geq t_2' \\ A_{0,1} D \{ \exp(-i\lambda_1(t_1 - t_2)) \} A_{0,1}^\dagger & \text{if } t_1 \geq t_1' \geq t_2 \geq t_2' \\ 0 & \text{otherwise} \end{array} \right\}$$

(23)

where $D\{ \cdot \}$ of a vector $v$ is a diagonal matrix with elements of $v$ on its diagonal, and the step from the first to second line again used $\exp(-iH_{\text{eff}}t)\ket{\phi} = \ket{\phi}$. To compute $g^{(2)}(t_1, t_2; \omega_L)$ as given by in Eq. 14a, we need to evaluate the integral:

$$\int_{t_1', t_2'}^{\infty} S_{c,c}(t_1, t_2; t_1', t_2') \exp[-i\omega_L(t_1' + t_2')] dt_1' dt_2'$$

(24)
In this section, we numerically verify that the transmissivity and two-photon-correlation expressions derived in the previous sections match with a master-equation based simulation of the multi-emitter CQED system. Figure 1 shows the comparison between master-equation based simulations (done using the open source python library QuTiP [3]) with the Scattering matrix approach for a two-emitter system. Within the master-equation framework, the coherent drive is incorporated by adding $\Omega(a + a^\dagger)$ to the system Hamiltonian, where $\Omega = \sqrt{\kappa_0 \beta_0}$ is the driving strength. We see that in the limit of $\beta_0$ (or $\Omega$) $\rightarrow$ 0, the QuTiP simulations agree perfectly with the scattering matrix based simulations, thereby validating the approach.

Next, we benchmark the scattering matrix approach — we compute the time taken to simulate the transmission and two-photon correlation through a system of up to 50 emitters. The results are shown in Fig. 2 — we note that even for a system of 50 emitters, computation of the two-photon correlation at a single frequency point takes up to 6s an 2.8 GHz Intel Core i7 processor with 16 GB RAM and 8 CPU cores, while utilizing ~1 CPU core. Hence, it would be possible to simulate many more emitters if desired. Moreover, we observe that the compute time for the transmission scales as $N^3$ and the compute time for the two-time correlation scales as $N^6$ — this is expected theoretically since their computation requires diagonalization of a matrix of size $\sim \mathcal{O}(N)$ and $\sim \mathcal{O}(N^2)$ respectively, and the compute time for diagonalization of a matrix of size $n$ scales as $n^3$. 

\section*{Validating and Benchmarking the Scattering Matrix Calculation}

Since the two-photon scattering matrix is symmetric with respect to an exchange of the indices $t_1'$ and $t_2'$, it follows that:

$$\int_{t_1',t_2'}^{\infty} S_{c,c}(t_1,t_2,t_1',t_2') \exp[-i\omega_L(t_1' + t_2')] dt_1'dt_2' = 2 \int_{t_1'=-\infty}^{t_1'} \int_{t_2'=-\infty}^{t_2'} S_{c,c}(t_1,t_2,t_1',t_2') \exp[-i\omega_L(t_1' + t_2')] dt_1'dt_2'$$

wherein the last step we have used the fact that if $t_1 \geq t_2$ and $t_1' \geq t_2'$, then the two photon scattering matrix $S_{c,c}(t_1,t_2,t_1',t_2')$ is 0 unless $t_2 \geq t_1'$ or $t_1 \geq t_1' \geq t_2 \geq t_2'$ (as shown in Eq. 23). These two integrals can be readily evaluated using Eq. 23 to obtain the following expression for the two-photon correlation function $g^{(2)}(t_1,t_2;\omega_L)$:

$$g^{(2)}(t_1,t_2;\omega_L) = \frac{\kappa_b\kappa_c}{T^2(\omega_L)} |g^T(\omega_L)1 + (f^T(\omega_L) - g^T(\omega_L)) \exp(-i(\lambda_1 - \omega_L)(t_1 - t_2))|^2$$

with $g(\omega_L)$ and $f(\omega_L)$ are defined by:

$$g(\omega_L) = d\{A_{0,1}D\left\{\frac{1}{\lambda_1 - \omega_L}\right\}A_{0,1}D\left\{\frac{1}{\lambda_1 - \omega_L}\right\}\}$$

$$f(\omega_L) = d\{A_{1,2}D\left\{\frac{1}{\lambda_2 - 2\omega_L}\right\}A_{1,2}D\left\{\frac{1}{\lambda_1 - \omega_L}\right\}A_{0,1}A_{0,1}\}$$

where $d\{\}$ of a square matrix $A$ is a vector with the diagonal elements of the matrix.
FIG. 2. Benchmarking the computation time of the scattering matrix approach with the number of emitters.

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