Theory of Frequency-Filtered and Time-Resolved N-Photon Correlations

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A theory of correlations between N photons of given frequencies and detected at given time delays is presented. These correlation functions are usually too cumbersome to be computed explicitly. We show that they are obtained exactly through intensity correlations between two-level sensors in the limit of their vanishing coupling to the system. This allows the computation of correlation functions hitherto unreachable. The uncertainties in time and frequency of the detection, which are necessary variables to describe the system, are intrinsic to the theory. We illustrate the power of our formalism with the example of the Jaynes-Cummings model, by showing how higher order photon correlations can bring new insights into the dynamics of open quantum systems.

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Photons emerged as a theoretical concept to explain fundamental properties of the electromagnetic field, such as the relationship between the energy of light and its frequency, thermal equilibrium of light and matter, or the photoelectric effect. With the advances in the generation, emission, transmission, and detection of photons, quantum systems are increasingly addressed at the single photon level and there is a pressing need for generalizations as well as refinements of the theory of photodetection [1]. For instance, photon correlations combining both their frequency and time information are now routinely measured in the laboratory. These experiments have proven extremely powerful in characterizing quantum systems such as a resonantly driven emitter [2–4], the strong coupling of light and matter [5–7], to perform quantum state tomography [8], to monitor heralded single photon sources [9], or to access spectral diffusion of single emitters [10].

At this level of fine control of the attributes of the quantum particles, one needs a theoretical description significantly more involved than general mathematical statements, such as the Wiener-Khinchin theorem which assumes abstract and unphysical properties of the light field. Eberly and Wódkiewicz, for instance, have shown how the physics of the detector needs to be included if a more realistic description of the light field is required [11]. In general, the more detailed is the characterization of a quantum system, the more necessary it becomes to describe its measurement. A bridge between the quantum system and the observer can be made with the so-called input-output formalism: the photons *inside* the system, say with operator a (we consider a single mode for simplicity), are weakly coupled to an *outside* continuum of modes, with operators A_ω (corresponding to their frequency ω). In the Heisenberg picture, the output field allows us to compute the time-dependent power spectrum of emission as the density of output photons with frequency ω_1 at time T_1, i.e., $S^{(1)}(\omega_1, T_1) = \langle A^\dagger_{\omega_1}(T_1)A_{\omega_1}(T_1)\rangle$. This quantity is physical only if the uncertainties of detection in both time and frequency are jointly taken into account [11]. Mathematically, this amounts to adding two exponential decays in the Fourier transform of the time autocorrelation $S^{(1)}_{\Gamma_1}(\omega_1, T_1) = \frac{\Gamma_1}{2\pi}\iint_{-\infty}^{T_1} dt_1' dt_4' e^{-(\Gamma_1/2)(T_1-t_1')}e^{-(\Gamma_1/2)(T_1-t_4')}\times e^{i\omega_1(t_4'-t_1')}\langle a^\dagger(t_1')a(t_4')\rangle$ where Γ_1 is interpreted as the linewidth of the detector. This so-called physical spectrum reduces to the Wiener-Khinchin theorem in the steady state and in the limit $\Gamma_1 \to 0$.

Extending this result for the detection of two photons was initially motivated by the Aspect *et al.* experiment [2] of resonance fluorescence in the Mollow triplet regime [12], where the peaks of the triplet were found to exhibit strong intensity correlations. These were described theoretically at first by dedicated methods for the problem at hand, from Cohen-Tannoudji *et al.* (dressed atom picture) [13,14] and Dalibard *et al.* (diagrammatic expansion)[15]. The extension of photodetection in the spirit of Eberly and Wódkiewicz by considering two detectors with respective linewidths Γ_1 and Γ_2 was impulsed by Knöll *et al.* [16] and Arnoldus and Nienhuis [17]. The expressions were of general validity, even though, due to their complexity, the authors still focused on the particular case of resonance fluorescence for illustration. The mathematical foundations, shaky in their initial development, were firmly established in the course of the following years [18–20]. The multiplicity of photons requires a careful time ($\mathcal{T}_\pm$) and normal (:) ordering of the operators [19,20], and it was realized that it is the time ordering of $\langle :A^\dagger_{\omega_1}(T_1)A_{\omega_1}(T_1)A^\dagger_{\omega_2}(T_2)A_{\omega_2}(T_2):\rangle$ which provides the physical two-photon spectrum $S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1, T_1; \omega_2, T_2) = \frac{\Gamma_1\Gamma_2}{(2\pi)^2}\iint_{-\infty}^{T_1} dt_1' dt_4' e^{-(\Gamma_1/2)(T_1-t_1')}e^{-(\Gamma_1/2)(T_1-t_4')}\iint_{-\infty}^{T_2} dt_2' dt_3'\times e^{-(\Gamma_2/2)(T_2-t_2')}e^{-(\Gamma_2/2)(T_2-t_3')}e^{i\omega_1(t_4'-t_1')}e^{i\omega_2(t_3'-t_2')}\langle\mathcal{T}_-[a^\dagger(t_1')\times a^\dagger(t_2')]\mathcal{T}_+[a(t_3')a(t_4')]\rangle$. Here, we have defined $\mathcal{T}_+$

(respectively $\mathcal{T}_-$) to order the operators in a product with the latest time to the far left (respectively, far right) [1]. Normalizing this expression yields the sought time- and frequency-resolved two-photon correlation function $g^{(2)}_{\Gamma_1\Gamma_2}(\omega_1, T_1; \omega_2, T_2) = S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1, T_1; \omega_2, T_2)/[S^{(1)}_{\Gamma_1}(\omega_1, T_1)S^{(1)}_{\Gamma_2}(\omega_2, T_2)]$. It is positive and finite, and reflects that frequency and time of emission cannot be both measured with arbitrary precision, in accordance with Heisenberg's uncertainty principle. The limiting behaviors of $g^{(2)}_{\Gamma_1\Gamma_2}$ defined in this way are those expected on physical grounds: photons are uncorrelated at infinite delays, $\lim_{|T_2-T_1|\to\infty} g^{(2)}_{\Gamma_1\Gamma_2}(\omega_1, T_1; \omega_2, T_2) = 1$ [21], and color-blind detectors recover the standard two-time correlators, $\lim_{\Gamma_1,\Gamma_2\to\infty} g^{(2)}_{\Gamma_1\Gamma_2}(\omega_1, T_1; \omega_2, T_2) = g^{(2)}(T_1; T_2)$. Further generalization to N-photon correlations follows in this way, adding pairs of operators with their corresponding integrals [18,22].

The actual computation of such $g^{(N)}_{\Gamma_1\ldots\Gamma_N}$, however, has proven so far to be intractable for $N > 2$, even for simple single-mode systems, such as resonance fluorescence or the single mode laser [23]. The case $N = 2$ is already demanding and thus some approximations were made to simplify the algebra [24,25]. More recently, the resonance fluorescence problem was revisited without approximations but still for two photons and at zero time delay only [26]. The main reason for such limitations is that all the possible time orderings of the $2N$-time correlator $\langle \mathcal{T}_-[a^\dagger(t_1')\ldots a^\dagger(t_N')]\mathcal{T}_+[a(t_{N+1}')\ldots a(t_{2N}')]\rangle$ result in $(2N-1)!!2^{N-1}$ independent terms. Furthermore, each of these correlators requires the application of the quantum regression theorem $2N-1$ times. This growth of the complexity makes a direct computation hopeless for a quantity which is otherwise straightforward to measure experimentally, merely by detecting photon clicks as a function of time and energy, a technology provided for instance by a streak camera [27].

In this Letter, we present a theory of N-photon correlations, that (i) allows for arbitrary time delays and frequencies, (ii) is applicable to any open quantum system and (iii) is both simple to implement and powerful. It consists in the introduction of N *sensors* to the dynamics of the open quantum system [noted Q in Fig. 1(a)]. Each sensor of the set $i = 1, \ldots, N$ is a two-level system with annihilation operator ς_i and transition frequency ω_i, that is matched to the frequency to be probed in the system. Its lifetime $1/\Gamma_i$ corresponds to the inverse detector linewidth. The coupling ε_i to each sensor is small enough so that the dynamics of the system is unaltered by their presence, with $\langle n_i\rangle = \langle \varsigma_i^\dagger \varsigma_i\rangle \ll 1$. More precisely, calling γ_Q any transition rate within Q (either with internal or external degrees of freedom) linked to the field of interest a, the tunnelling rates ε_i must be such that losses into the sensors and their back action are negligible, leading to $\varepsilon_i \ll \sqrt{\Gamma_i\gamma_Q/2}$. Under this condition, we solve the full quantum dynamics of the system supplemented with the N sensors. The latter play the role of the output fields $A_{\omega_i}(t)$, but instead of formally solving the Heisenberg equations and expressing their correlations in terms of the system operators (as in the standard method exposed above), we compute directly intensity-intensity correlations between sensors, which is a considerably simpler task. The main result of this Letter, which is demonstrated in the Supplemental Material [28], is:

$$g^{(N)}_{\Gamma_1\ldots\Gamma_N}(\omega_1, T_1; \ldots; \omega_N, T_N) = \lim_{\varepsilon_1,\ldots,\varepsilon_N\to 0} \frac{\langle n_1(T_1)\ldots n_N(T_N)\rangle}{\langle n_1(T_1)\rangle\ldots\langle n_N(T_N)\rangle}, \quad (1)$$

where the left-hand side is the time- and frequency-resolved N-photon correlation function as defined previously [29]. The Supplemental Material [28] establishes that, for open quantum systems described by Lindblad type master equations, $\langle n_1(T_1)\ldots n_N(T_N)\rangle = \frac{\varepsilon_1^2\ldots\varepsilon_N^2}{\Gamma_1\ldots\Gamma_N}(2\pi)^N S^{(N)}_{\Gamma_1\ldots\Gamma_N}(\omega_1, T_1; \ldots; \omega_N, T_N)$ to leading order in the ε_i, which proves Eq. (1). The equality is of general validity with no approximations or assumptions on the system. With this result, the complexity of computing $g^{(N)}_{\Gamma_1\ldots\Gamma_N}(\omega_1, T_1; \ldots; \omega_N, T_N)$ is greatly reduced as no integral needs to be computed and the quantum regression theorem needs to be applied $N-1$ times only. For the important case of zero delay, $g^{(N)}_{\Gamma_1\ldots\Gamma_N}(\omega_1; \ldots; \omega_N)$ reduces to a single-time averaged quantity. N degenerate sensors with frequency ω and linewidth Γ also provide the Nth-order correlations of a single harmonic oscillator with frequency ω and linewidth Γ, corresponding to the case of correlations measured after the application of a single filter. This method is also useful to derive analytical results (as shown in the Supplemental Material [28]).

We now illustrate its efficiency and ease of use by applying it to the Jaynes-Cummings model [30], which is both an important and fundamental quantum description of light-matter interaction [31], is much more complex than resonance fluorescence as it also quantizes the light field [32], and is particularly suited to generate strongly correlated photons [33,34]. Our method recovers exactly the known results for the Mollow triplet [24–26], and extends them effortlessly.

At resonance between the light mode (a) and the two-level emitter (σ) both with bare frequency ω_a, the Jaynes-Cummings Hamiltonian reads $H = g(a^\dagger\sigma + a\sigma^\dagger)$. The master equation that describes decay (γ_a, γ_σ) and incoherent pumping of the emitter (P_σ) has the form $\partial_t\rho = i[\rho, H] + [\frac{\gamma_a}{2}\mathcal{L}_a + \frac{\gamma_\sigma}{2}\mathcal{L}_\sigma + \frac{P_\sigma}{2}\mathcal{L}_{\sigma^\dagger}](\rho)$, where $\mathcal{L}_c(O) = (2cOc^\dagger - c^\dagger cO - Oc^\dagger c)$ and ρ is the density matrix for the emitter and cavity system [35]. The new density matrix that includes the sensors, ρ_{sen}, follows a modified master equation where the photonic tunnelling

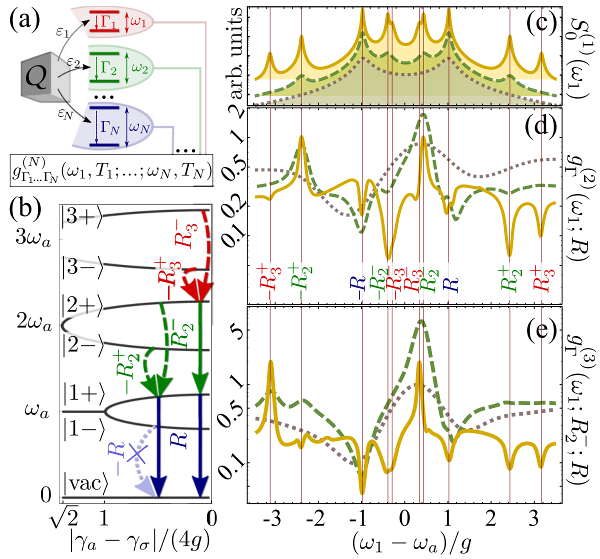

FIG. 1 (color online). (a) Scheme of our proposal to compute N-photon correlations between photons emitted at different times and frequencies from an open quantum open system Q. N two-level systems of ascribed frequencies are weakly coupled to Q and serve as correlation sensors at these frequencies, with their decay rate providing the detector linewidth. (b) Dissipative Jaynes-Cummings ladder up to the third rung with two of the cascades probed in panels (d) [with two sensors] and (e) [with three sensors]. Solid arrows show the fixed frequencies. Curved arrows show the scanning frequency ω_1, at the transitions where the joint emission is strongly enhanced (dashed) or, on the other hand, suppressed (dotted). (c) Power spectra of emission probed by weak incoherent excitation ($P_\sigma = \gamma_\sigma = 0.01g$) for three cavities of decreasing quality $\gamma_a = 0.01$ (solid), 0.1 (dashed) and $0.5g$ (dotted). (d) Two- and (e) three-photon correlations at zero delay for the three cavities, with sensor linewidths $\Gamma = \gamma_2$ (solid) and $\gamma_2/2$ (dashed and dotted).

terms, $H_{\text{sen}} = \sum_{i=1}^{N}[\omega_i \varsigma_i^\dagger \varsigma_i + \varepsilon_i(a\varsigma_i^\dagger + a^\dagger \varsigma_i)]$, are added to the original Hamiltonian, and the sensor decay terms $\sum_{i=1}^{N} \frac{\Gamma_i}{2}\mathcal{L}_{\varsigma_i}(\rho_{\text{sen}})$ are added to the dissipative part. The level structure of the dressed states $|n, \pm\rangle$ with n excitations is given by the dissipative Jaynes-Cummings ladder [35], which is shown in Fig. 1(b) at low pumping, $P_\sigma = \gamma_\sigma$, and in the strong-coupling regime with $\gamma_\sigma \leq \gamma_a < 4g$. This gives rise to the transition frequencies $R_n^\pm = \sqrt{ng^2 - (\frac{\gamma_a - \gamma_\sigma}{4})^2} \pm \sqrt{(n-1)g^2 - (\frac{\gamma_a - \gamma_\sigma}{4})^2}$ between rungs for $n \geq 2$ with broadening $\gamma_n = 2(n-1)\gamma_a + \gamma_\sigma$ [35]. The Rabi splitting $2R$, which arises from transitions $|1\pm\rangle \to |\text{vac}\rangle$, is given by $R = \sqrt{g^2 - (\frac{\gamma_a - \gamma_\sigma}{4})^2}$ with $\gamma_1 = (\gamma_a + \gamma_\sigma)/2$. These transitions result in peaks in the power spectrum, as seen in Fig. 1(c) for the three cavity decay rates $\gamma_a/g = 0.01$, 0.1, and 0.5 that are chosen to correspond to cavities embedding superconducting qubits [36], atoms [37], and quantum dots [38], respectively. They all show the first rung transitions at $\pm R$, the so-called Rabi doublet, and one can distinguish outer peaks at $\pm R_n^+$ and inner peaks at $\pm R_n^-$, up to the third rung for the best system (solid line) and to the second rung for the intermediate one (dashed line). In Fig. 1(d), we set the linewidth of the sensors Γ at a value around γ_2 and compute the two-photon correlation at zero delay, $g_\Gamma^{(2)}(\omega_1; \omega_2)$, between a photon with fixed frequency at the Rabi peak, $\omega_2 = R$ [solid arrow on the left of Fig. 1(b)], and a photon with variable frequency ω_1 which scans the spectral range (curved arrows). When the scanning frequency ω_1 matches the second rung transitions that are precursors of the Rabi transition R, the probability of joint emission is enhanced relatively to other frequencies. The filtering then tracks photons in the cascades $|2+\rangle \to |1+\rangle$ at $\omega_1 = R_2^-$ and $|2-\rangle \to |1+\rangle$ at $-R_2^+$. This is a common feature to all three systems, which shows that even if broadening is too large to observe explicit features from higher rungs in the power spectrum, $g_\Gamma^{(2)}(\omega_1; \omega_2)$ allows us to uncover them in the photon correlations. On the other hand, we obtain the expected strong suppression when the first photon is detected at the other branch of the Rabi doublet, $\omega_1 = -R$. More features can be observed for the better systems such as dips at the two remaining transitions from the second rung, $|2-\rangle \to |1-\rangle$ at $\omega_1 = -R_2^-$ and $|2+\rangle \to |1-\rangle$ at R_2^+. In the best system, we can even resolve the dips for the third rung transitions at $\omega_1 = \pm R_3^\pm$. All these transitions do not form a consecutive cascade with the one we fixed and therefore have less probability to occur within the considered small time window $1/\gamma_2$.

Instead of making a comprehensive analysis of $g_\Gamma^{(2)}$ specifics, we now turn to higher order correlation functions, such as the simultaneous three-photon correlations $g_\Gamma^{(3)}(\omega_1; \omega_2; \omega_3)$, which are exceedingly hard to compute with previous methods. We fix two frequencies of detection at $\omega_2 = R_2^-$ and $\omega_3 = R$ [solid arrows on the right of Fig. 1(b)] and again let ω_1 vary. A strong enhancement is also observed for all systems, now at $\omega_1 = R_3^-$ which monitors the cascade $|3+\rangle \to |2+\rangle \to |1+\rangle \to |\text{vac}\rangle$ depicted in Fig. 1(b) and at $\omega_1 = -R_3^+$ which starts it with $|3-\rangle \to |2+\rangle$. Other transitions show dips that are also clearly understood. This hints at the possible characterization of the level structure of an open quantum system. In general, however, one cannot draw conclusions from the zero-delay case only, in particular for small features, such as the small enhancement at $\omega_1 = -R_2^+$ in $g_\Gamma^{(3)}$ (for the dashed line only) which is not necessarily a bunching peak and reveals itself in the τ dynamics to be antibunched, as discussed later.

In Fig. 2(a), we explore another important aspect of $g_\Gamma^{(N)}$, namely the dependence of correlations on the sensors linewidths, which is related to the complementary uncertainties in time and frequency. In the case $\Gamma \to 0$ of perfect detectors, $g_0^{(N)} = 1$ for all N with nondegenerate frequencies, since the complete indeterminacy in time leads to averaging photons from all possible time delays. For M

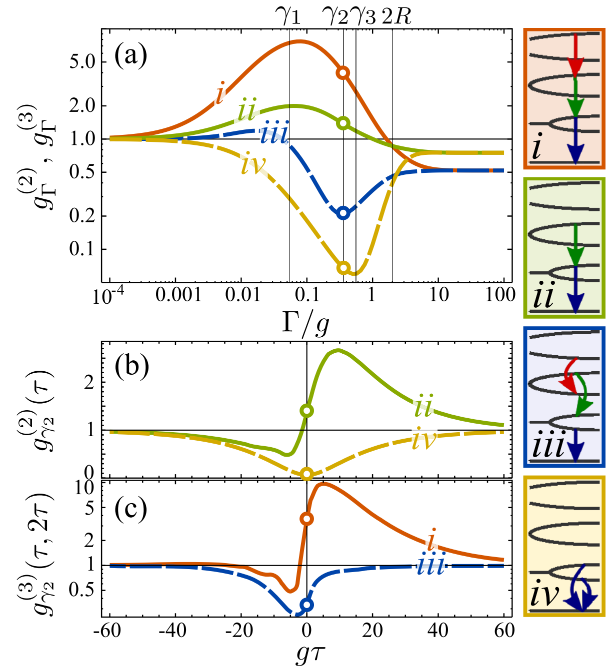

FIG. 2 (color online). (a) Two- and three-photon correlations at zero delay as a function of the sensor linewidth Γ, with frequencies of detection as shown in the insets *i*–*iv*. (b)–(c) τ dynamics of the correlation functions with $\Gamma = \gamma_2$ for, (b), two photons in the configurations of insets *ii* and *iv* and, (c), three photons in the configurations *i* and *iii*. Positive τ corresponds to detection in the order from top to bottom of the ladder. Parameters: $P_\sigma = \gamma_\sigma = 0.01g$, $\gamma_a = 0.1g$.

degenerate frequencies out of N, photon indistinguishability results in $M!$ ways for the sensors to measure the same configuration, that is, $\lim_{\Gamma\to 0} g_\Gamma^{(N)} = M!$. This limit has been misunderstood in the literature [39]. The effect has otherwise been reported for the case $M = N = 2$ by converting laser light into chaotic light with narrow filters [23]. The other limit $\Gamma \to \infty$ corresponds to the opposite situation of exact τ delay between photons of completely indeterminate frequencies. This is of more interest, in particular at zero time delay, which is the case of Fig. 2(a). For the Jaynes-Cummings system at low pumping, this recovers results derived by other approaches [40,41].

The intermediate case of finite linewidth of the sensors is the most interesting. Features are the most marked when detector linewidths are of the order of those of the transitions involved, since the peaks of the spectrum are best filtered. Smaller linewidths (longer times) are to be favored for bunching and larger linewidths (smaller times) for antibunching. One sees, for instance in Fig. 2(a), that consecutive transitions, forming a cascade—such as those sketched in panel *i* (with three photons) or *ii* (with two photons)—show an enhancement. Conversely, the simultaneous emission from both Rabi peaks, in the configuration sketched as *iv*, is substantially suppressed, leading to strong antibunching. This observation with a microcavity containing a single quantum dot has been used to demonstrate the quantum nature of strong light-matter coupling [6] (with detuning to better separate the peaks). Further theoretical investigations with this formalism (to be discussed elsewhere) may help to elucidate the nature of spectral triplets also observed in such experiments [6,42,43].

Figures 2(b) and 2(c) show an example of the τ dependence of the correlations, for the case $\Gamma = \gamma_2$, both at positive and negative delays. The configuration *ii* has the typical shape of a cascade between consecutive levels, with antibunching for $\tau < 0$, a step at $\tau = 0$, and bunching for $\tau > 0$. This behavior is well known, for instance from the biexciton-exciton cascade [9]. It is also observed for N photons in any consecutive transitions, such as is shown in *i* for three photons starting from the third rung. In contrast, the filtering of peaks which do not belong to the same cascade exhibit antibunching, as seen in *iv* for the two Rabi peaks or *iii* for one of its three-photon counterparts: the order of the transition does not matter anyway and the cases $\pm\tau$ show qualitatively the same behavior. These results are, to the best of our knowledge, the first computations of three-time frequency-resolved correlation functions. They are easily extended to higher orders (a fourth order example is given in the Supplemental Material [28]).

In conclusion, we have presented a theory to efficiently compute correlations between an arbitrary number of photons of any given frequencies and time delays. All three aspects of the detection, namely frequencies, time delays, and linewidths of the detectors, are needed to characterize meaningfully the system. The method allows us to compute exactly, with low effort and for general open quantum systems, properties of output fields that are otherwise defined in terms of complicated integrals. Its ease of use enabled us to present the first computation of three and four time-resolved and frequency-filtered correlation functions. Its application will allow the interpretation of experiments which are routinely implemented in the laboratory but which lacked hitherto an adequate and tractable theoretical support, and to design new ways to unravel and/or engineer the quantum dynamics of open systems.

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[28] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.109.183601 for the proof of the equivalence between the sensor and the integral methods and for further illustrations (up to four-photon correlations).
[29] Its explicit integral form is given for the case $N = 2$ in the Supplemental Material [28], cf. Eq. (17) normalized by $S^{(1)}_{\Gamma_1}(\omega_1, T_1)S^{(1)}_{\Gamma_2}(\omega_2, T_2)$.
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Supplemental Material of "Theory of Frequency-Filtered and Time-Resolved N-Photon Correlations"

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We prove the equality between the experimentally motivated correlation function $g^{(N)}_{\Gamma_1\dots\Gamma_N}(\omega_1,T_1;\dots;\omega_N,T_N)$—defined as $2N$-time integrals—and the intensity correlation $\langle n_1(T_1)\dots n_N(T_N)\rangle/(\langle n_1(T_1)\rangle\dots\langle n_N(T_N)\rangle)$ between sensors coupled to the system, to leading order in the coupling. We also further illustrate the method by calculating correlations in the Jaynes–Cummings model up to fourth order.

PROOF OF THE EQUIVALENCE BETWEEN THE SENSING AND THE INTEGRAL METHODS

Let us assume a quantum system described by a set of operators a, σ, etc., acting in a Hilbert space $\mathcal{H}$. In second quantization, these operators define annihilation operators in the Heisenberg picture. The system can be fully Bosonic, Fermionic or a mixture involving any number of operators. All single-time quantities can be obtained from correlators of the type $\langle a^{\dagger\mu}a^{\nu}\sigma^{\dagger\eta}\sigma^{\theta}\dots\rangle$ with μ, ν, η, θ, etc., integers. Let us call $\mathcal{O}$ the set of operators the averages of which correspond to the correlators required to describe the system, i.e., $\mathcal{O}$ includes all the sought observables as well as operators which couple to them through the equations of motion. In the following, we assume, without loss of generality, that a is the mode of interest, the correlations of which are to be computed in time and frequency.

We prove the case $N=1$ first, which corresponds to the power spectrum, then $N=2$, which corresponds to the most important correlation function. The proof admits a straightforward generalization to higher N. The proof proceeds by computing separately the integral expressions on the one hand and the intensity correlations between sensors on the other hand, and showing that they are equal to leading order in the couplings to the sensors. We assume the steady state case for simplicity, with little loss of generality.

$N=1$, power spectrum

Integral method

The single-photon physical spectrum introduced in the main text as $S^{(1)}_{\Gamma_1}(\omega_1,T_1)=\frac{\Gamma_1}{2\pi}\times\iint_{-\infty}^{T_1}dt_1'dt_4'e^{-\frac{\Gamma_1}{2}(T_1-t_1')}e^{-\frac{\Gamma_1}{2}(T_1-t_4')}e^{i\omega_1(t_4'-t_1')}\langle a^\dagger(t_1')a(t_4')\rangle$ can, through convolutions, be put in the form of an uncertainty in the time of detection [1]:

$$S^{(1)}_{\Gamma_1}(\omega_1,T_1)=\Gamma_1\int_{-\infty}^{T_1}dt_1e^{-\Gamma_1(T_1-t_1)}\Sigma^{(1)}_{\Gamma_1}(\omega_1,t_1)\qquad(1)$$

where

$$\Sigma^{(1)}_{\Gamma_1}(\omega_1,t_1)=\frac{1}{\pi}\Re\int_0^\infty d\tau_1e^{-\frac{\Gamma_1}{2}\tau_1}e^{-i\omega_1\tau_1}\langle a^\dagger(t_1)a(t_1-\tau_1)\rangle\qquad(2)$$

contains the uncertainty in the frequency of detection [2]: $\Sigma^{(1)}_{\Gamma_1}(\omega_1,t_1)=\int_{-\infty}^{\infty}d\omega_1'\Sigma^{(1)}_0(\omega_1',t_1)\frac{1}{\pi}\frac{\frac{\Gamma_1}{2}}{(\frac{\Gamma_1}{2})^2+(\omega_1'-\omega_1)^2}$. The kernel of this expression corresponds to the case of a perfect detector, $\Gamma_1=0$, known as the Page–Lampard quasi-spectrum of emission $\Sigma^{(1)}_0(\omega_1,t_1)$ [3]. The results of Eberly and Wódkiewicz [1] show that the time-dependent physical spectrum (1) is *i*) always positive, whereas $\Sigma^{(1)}_0(\omega_1,t_1)$ is not in general, and *ii*) finite, even in the steady state $S^{(1)}_{\Gamma_1}(\omega_1,T_1\to\infty)=\Sigma^{(1)}_{\Gamma_1}(\omega_1,t_1)$, whereas $S^{(1)}(\omega_1,T_1\to\infty)$ diverges.

To compute Eq. (1), we only need to obtain the two-time correlator $\langle a^\dagger(t_1)a(t_1-\tau_1)\rangle$. For any two operators X and Y acting on $\mathcal{H}$, we define the vector $\mathbf{v}_{X,Y}(\tau)$ as:

$$\mathbf{v}_{X,Y}(\tau)=\begin{pmatrix}\langle X(0)Y(0)\rangle\\ \langle X(0)a(\tau)Y(0)\rangle\\ \langle X(0)a^\dagger(\tau)Y(0)\rangle\\ \langle X(0)(a^\dagger a)(\tau)Y(0)\rangle\\ \vdots\end{pmatrix},\qquad(3)$$

where X and Y, in the steady state, sandwich the operators of $\mathcal{O}$ taken in some order, which will be kept for the remainder of the text as starting with the sequence $\mathcal{O}=\{1,a,a^\dagger,a^\dagger a,\dots\}$.

From the quantum regression theorem, one can define for $\mathcal{O}$ a matrix M which rules the dynamical evolution of $\mathbf{v}_{X,Y}$:

$$\partial_\tau\mathbf{v}_{X,Y}(\tau)=M\mathbf{v}_{X,Y}(\tau)\,,\qquad(4)$$

with solution $\mathbf{v}_{X,Y}(\tau)=e^{M\tau}\mathbf{v}_{X,Y}(0)$. The steady state of the system is then fully given by:

$$\mathbf{v}^{\mathrm{ss}} = \lim_{\tau\to\infty} \mathbf{v}_{1,1}(\tau) = \lim_{\tau\to\infty} e^{M\tau} \begin{pmatrix} 1 \\ 0 \\ \vdots \end{pmatrix} , \quad (5)$$

since $\mathcal{O}$ contains all the relevant observables of the system. Here we have chosen the vacuum as the initial condition. Since we employ the standard assumption of a unique steady state, the initial state does not matter and all the information is encoded in $e^{M\tau}$.

We now define two matrices, $T_\pm$, which, when acting on $\mathbf{v}_{X,Y}(\tau)$, introduce an extra $a^\dagger$ for T_+ and an a for T_- between X and Y, keeping normal ordering:

$$T_+\mathbf{v}_{X,Y}(\tau) = \begin{pmatrix} \langle X(0)a^\dagger(\tau)Y(0)\rangle \\ \langle X(0)(a^\dagger a)(\tau)Y(0)\rangle \\ \langle X(0)a^{\dagger 2}(\tau)Y(0)\rangle \\ \langle X(0)(a^{\dagger 2}a)(\tau)Y(0)\rangle \\ \vdots \end{pmatrix} , \quad (6)$$

and

$$T_-\mathbf{v}_{X,Y}(\tau) = \begin{pmatrix} \langle X(0)a(\tau)Y(0)\rangle \\ \langle X(0)a^2(\tau)Y(0)\rangle \\ \langle X(0)(a^\dagger a)(\tau)Y(0)\rangle \\ \langle X(0)(a^\dagger a^2)(\tau)Y(0)\rangle \\ \vdots \end{pmatrix} . \quad (7)$$

These matrices always exist, in infinite or in truncated Hilbert spaces (where, if truncation is to order n, a^n is an operator in $\mathcal{O}$ but $a^{n+1}=0$). For instance, if the mode a is a two-level system, the vector $\mathbf{v}_{X,Y}(\tau)$ consists of the first four entries in Eq. (3) only, since $a^{\dagger\mu}a^\nu = 0$ if μ or $\nu > 1$. Then, these matrices read

$$T_+ = \begin{pmatrix} 0&0&1&0 \\ 0&0&0&1 \\ 0&0&0&0 \\ 0&0&0&0 \end{pmatrix} \quad \text{and} \quad T_- = \begin{pmatrix} 0&1&0&0 \\ 0&0&0&0 \\ 0&0&0&1 \\ 0&0&0&0 \end{pmatrix} . \quad (8)$$

With these definitions, the correlator $\langle a^\dagger(t_1)a(t_1-\tau_1)\rangle$ with $\tau_1>0$ is the first element of $T_+\mathbf{v}_{1,a}(\tau_1)$:

$$\langle a^\dagger(t_1)a(t_1-\tau_1)\rangle = \left[T_+e^{M\tau_1}T_-\mathbf{v}^{\mathrm{ss}}\right]_1 , \quad (9)$$

where we have used $[\cdots]_i$ to denote the ith element of a vector. The power spectrum in its integral form is therefore given by:

$$S^{(1)}_{\Gamma_1}(\omega_1) = \frac{1}{\pi}\Re\left[T_+\frac{-1}{M+(-i\omega_1-\frac{\Gamma_1}{2})\mathbf{1}}T_-\mathbf{v}^{\mathrm{ss}}\right]_1 , \quad (10)$$

with $\mathbf{1}$ the identity matrix.

Sensing method

We now consider two sensors ς_i, $i=1,2$ with linewidths Γ_i coupled to the system with strength ε_i such that the dynamics of the system is probed but is otherwise left unperturbed. This requires the tunnelling rates ε_i to fulfil two conditions: the losses into the sensors must be negligible, $4\varepsilon_i^2/\Gamma_i \ll \gamma_Q$ and so must be the back action of the sensors into the system, $4\varepsilon_i^2/\gamma_Q \ll \Gamma_i$, where γ_Q is the smallest system decay rate. These conditions both lead to $\varepsilon_i \ll \sqrt{\Gamma_i\gamma_Q/2}$. We then introduce a sensing vector $\mathbf{w}$ of steady state correlators, by multiplying $\varsigma_1^{\dagger\mu_1}\varsigma_1^{\nu_1}\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}$ with the operators in $\mathcal{O}$:

$$\mathbf{w}[\mu_1\nu_1,\mu_2\nu_2] = \begin{pmatrix} \langle\varsigma_1^{\dagger\mu_1}\varsigma_1^{\nu_1}\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}\rangle \\ \langle\varsigma_1^{\dagger\mu_1}\varsigma_1^{\nu_1}\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}a\rangle \\ \langle\varsigma_1^{\dagger\mu_1}\varsigma_1^{\nu_1}\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}a^\dagger\rangle \\ \langle\varsigma_1^{\dagger\mu_1}\varsigma_1^{\nu_1}\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}a^\dagger a\rangle \\ \vdots \end{pmatrix} , \quad (11)$$

where the indices μ_i and ν_i take the values 0 or 1. In the regime under consideration, the population $\langle\varsigma_i^\dagger\varsigma_i\rangle \ll 1$ and the equations of motion are valid to leading order in $\varepsilon_{1,2}$:

$$\begin{aligned}\partial_t\mathbf{w}[\mu_1\nu_1,\mu_2\nu_2] = {}& \{M + [(\mu_1-\nu_1)i\omega_1 - (\mu_1+\nu_1)\frac{\Gamma_1}{2} + (\mu_2-\nu_2)i\omega_2 - (\mu_2+\nu_2)\frac{\Gamma_2}{2}]\mathbf{1}\}\mathbf{w}[\mu_1\nu_1,\mu_2\nu_2] \\ &+ \mu_1(i\varepsilon_1T_+)\mathbf{w}[0\nu_1,\mu_2\nu_2] + \nu_1(-i\varepsilon_1T_-)\mathbf{w}[\mu_10,\mu_2\nu_2] + \mu_2(i\varepsilon_2T_+)\mathbf{w}[\mu_1\nu_1,0\nu_2] + \nu_2(-i\varepsilon_2T_-)\mathbf{w}[\mu_1\nu_1,\mu_20] , \quad (12)\end{aligned}$$

and can be solved recursively:

$$\begin{aligned}\mathbf{w}[\mu_1\nu_1,\mu_2\nu_2] = {}& \frac{-1}{M + [(\mu_1-\nu_1)i\omega_1 - (\mu_1+\nu_1)\frac{\Gamma_1}{2} + (\mu_2-\nu_2)i\omega_2 - (\mu_2+\nu_2)\frac{\Gamma_2}{2}]\mathbf{1}} \\ &\times\Big\{\mu_1(i\varepsilon_1T_+)\mathbf{w}[0\nu_1,\mu_2\nu_2] + \nu_1(-i\varepsilon_1T_-)\mathbf{w}[\mu_10,\mu_2\nu_2] + \mu_2(i\varepsilon_2T_+)\mathbf{w}[\mu_1\nu_1,0\nu_2] + \nu_2(-i\varepsilon_2T_-)\mathbf{w}[\mu_1\nu_1,\mu_20]\Big\} . \quad (13)\end{aligned}$$

Higher order terms will cancel exactly in the vanishing coupling we will assume later and thus do not need to be included here. Besides, unlike the leading order term, the higher order ones depend on the modelling of the sensors (as two-level systems, harmonic oscillators, etc.) and on the system itself.

The spectrum of emission of a is given by the average population, in the steady state, of any one of the two sensors, say, the first one: $\langle n_1\rangle = \langle\varsigma_1^\dagger\varsigma_1\rangle$. Its equation of motion reads $\partial_t\langle n_1\rangle = -\Gamma_1\langle n_1\rangle + 2\Re(i\varepsilon_1\langle\varsigma_1 a^\dagger\rangle)$, and with the above notations, is therefore given in the steady state by:

$$\langle n_1\rangle = \frac{2}{\Gamma_1}\Re\Big[i\varepsilon_1 T_+\mathbf{w}[01,00]\Big]_1\,. \tag{14}$$

Using the solution Eq. (13), the correlator of interest for the spectrum reads:

$$\mathbf{w}[01,0,0] = \frac{-1}{M+[-i\omega_1-\frac{\Gamma_1}{2}]\mathbf{1}}(-i\varepsilon_1 T_-)\mathbf{v}^{\mathrm{ss}}\,. \tag{15}$$

Equality of the integral and sensing methods

The proof is now complete since, to leading order, we find that Eq. (10) and Eqs. (14-15) provide the claimed identity:

$$\langle n_1\rangle = \frac{2\varepsilon_1^2}{\Gamma_1}\Re\left[T_+\frac{-1}{M+[-i\omega_1-\frac{\Gamma_1}{2}]\mathbf{1}}T_-\mathbf{v}^{\mathrm{ss}}\right]_1$$
$$= \frac{\varepsilon_1^2}{\Gamma_1}(2\pi)S^{(1)}_{\Gamma_1}(\omega_1)\,. \tag{16}$$

$N=2$, two-photon correlations

Integral method

The case $N=2$ brings with the multiplicity of photons the conceptual difficulty of time- and normal-ordering. It was discussed in the text that the proper definition yielding a physical two-photon spectrum reads [4, 5]:

$$S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1,T_1;\omega_2,T_2) = \frac{\Gamma_1\Gamma_2}{(2\pi)^2}\iint_{-\infty}^{T_1}dt_1'dt_4'e^{-\frac{\Gamma_1}{2}(T_1-t_1')}e^{-\frac{\Gamma_1}{2}(T_1-t_4')}\iint_{-\infty}^{T_2}dt_2'dt_3'e^{-\frac{\Gamma_2}{2}(T_2-t_2')}e^{-\frac{\Gamma_2}{2}(T_2-t_3')}$$
$$\times\, e^{i\omega_1(t_4'-t_1')}e^{i\omega_2(t_3'-t_2')}\langle\mathcal{T}_-[a^\dagger(t_1')a^\dagger(t_2')]\mathcal{T}_+[a(t_3')a(t_4')]\rangle\,. \tag{17}$$

In analogy with the case $N=1$, it can be put in the form:

$$S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1,T_1;\omega_2,T_2) = \Gamma_1\Gamma_2\int_{-\infty}^{T_1}dt_1\int_{-\infty}^{T_2}dt_2 e^{-\Gamma_1(T_1-t_1)}e^{-\Gamma_2(T_2-t_2)}\Sigma^{(2)}_{\Gamma_1\Gamma_2}(\omega_1,t_1;\omega_2,t_2)\,, \tag{18}$$

isolating the *two-photon quasi-distribution*:

$$\Sigma^{(2)}_{\Gamma_1\Gamma_2}(\omega_1,t_1;\omega_2,t_2) = \frac{2\Re}{(2\pi)^2}\iint_0^\infty d\tau_1 d\tau_2 e^{-\frac{\Gamma_1}{2}\tau_1}e^{-\frac{\Gamma_2}{2}\tau_2}$$
$$\times\, e^{-i\omega_2\tau_2}[e^{i\omega_1\tau_1}\langle\mathcal{T}_-[a^\dagger(t_1-\tau_1)a^\dagger(t_2)]\mathcal{T}_+[a(t_2-\tau_2)a(t_1)]\rangle + e^{-i\omega_1\tau_1}\langle\mathcal{T}_-[a^\dagger(t_1)a^\dagger(t_2)]\mathcal{T}_+[a(t_2-\tau_2)a(t_1-\tau_1)]\rangle]\,, \tag{19}$$

which, like the quasi-spectrum, can be negative and is thus not a physical spectrum.

To proceed with the calculation, let us separate the $\tau = T_2-T_1$ two-photon correlation function between its $\tau=0$ and $\tau>0$ terms:

$$S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2,\tau) =$$
$$e^{-\Gamma_2\tau}S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2) + \Delta S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2,\tau)\,, \tag{20}$$

with

$$S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2) = \Gamma_1\Gamma_2\int_{-\infty}^{T_1}dt_2\int_{-\infty}^{t_2}dt_1$$
$$\times e^{-\Gamma_1(T_1-t_1)}e^{-\Gamma_2(T_1-t_2)}\Sigma^{(2)}_{\Gamma_1\Gamma_2}(\omega_1,t_1;\omega_2,t_2)+[1\leftrightarrow 2]\,, \tag{21}$$

and

$$\Delta S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2,\tau) = \Gamma_1\Gamma_2\int_{T_1}^{T_2}dt_2\int_{-\infty}^{T_1}dt_1$$
$$\times\, e^{-\Gamma_1(T_1-t_1)}e^{-\Gamma_2(T_2-t_2)}\Sigma^{(2)}_{\Gamma_1\Gamma_2}(\omega_1,t_1;\omega_2,t_2)\,, \tag{22}$$

where $[1\leftrightarrow 2]$ means the interchange of sensors 1 and 2, that is, permuting $\omega_1\leftrightarrow\omega_2$ and $\Gamma_1\leftrightarrow\Gamma_2$.

To compute these quantities, it is enough to consider $\Sigma^{(2)}_{\Gamma_1\Gamma_2}(\omega_1,t_1;\omega_2,t_2)$ for $t=t_2-t_1>0$ since the inverse order is given by the exchange $[1\leftrightarrow 2]$. Therefore, we restrict the integration to ordering of the time variables where $t_1-\tau_1<t_1<t_2$. The fourth variable yields three different domains of integration:

(1) $t_2-\tau_2<t_1-\tau_1<t_1<t_2$,
(2) $t_1-\tau_1<t_2-\tau_2<t_1<t_2$,
(3) $t_1-\tau_1<t_1<t_2-\tau_2<t_2$.

For each of them, there are two different correlators appearing in $\Sigma^{(2)}$: one with the factor $e^{-i\omega_2\tau_2}e^{i\omega_1\tau_1}$, the other with $e^{-i\omega_2\tau_2}e^{-i\omega_1\tau_1}$. They will be respectively referred to as $\mathcal{C}_{(ia)}$ and $\mathcal{C}_{(ib)}$, with $i=1,2,3$ depending on their domains of integration. This gives rise to six integrals which we shall denote $\mathcal{I}_{(ia)}$ and $\mathcal{I}_{(ib)}$.

From this discussion, we can find a general expression for the complexity of the integration method in terms of the various domains of integration and the different correlators to be considered. The number of independent time ordering is $(2N-1)!!$ and the number of independent terms in $\Sigma^{(N)}$ is $2^N/2$ (we divide by 2 because half are complex conjugates of the other half). The total number of independent time integrals and correlators is therefore $(2N-1)!!2^{N-1}$.

The first correlator we need, $\mathcal{C}_{(1a)}=\langle a^\dagger(t_1-\tau_1)a^\dagger(t_2)a(t_1)a(t_2-\tau_2)\rangle$, is the first element of the vector $T_+\mathbf{v}_{X_1,Y_1}(t)$ with $X_1=a^\dagger(t_1-\tau_1)$ and $Y_1=a(t_1)a(t_2-\tau_2)$. We obtain $\mathbf{v}_{X_1,Y_1}(t)=e^{Mt}\mathbf{v}_{X_1,Y_1}(0)=e^{Mt}T_-\mathbf{v}_{X_1,Y_2}(\tau_1)$ with $Y_2=a(t_2-\tau_2)$. In turn, $\mathbf{v}_{X_1,Y_2}(\tau_1)=e^{M\tau_1}\mathbf{v}_{X_1,Y_2}(0)=e^{M\tau_1}T_+\mathbf{v}_{1,Y_2}(t')$ is obtained with $Y_2=a(t_2-\tau_2)$ and $t'=\tau_2-\tau_1-t$. Finally, we get $\mathbf{v}_{1,Y_2}(t')=e^{Mt'}\mathbf{v}_{1,Y_2}(0)=e^{Mt'}T_-\mathbf{v}^{\mathrm{ss}}$. Putting everything together, we get:

$$\mathcal{C}_{\substack{(1a)\\(1b)}}=\Big[T_+e^{Mt}T_\mp e^{M\tau_1}T_\pm e^{Mt'}T_-\mathbf{v}^{\mathrm{ss}}\Big]_1\,, \tag{23}$$

with correspondence between upper and lower indices with the sign. Repeating this procedure for the other domains of integration, we also get:

$$\mathcal{C}_{\substack{(2a)\\(2b)}}=\Big[T_+e^{Mt}T_\mp e^{-Mt}e^{M\tau_2}T_-e^{Mt''}T_\pm\mathbf{v}^{\mathrm{ss}}\Big]_1\,, \tag{24}$$

where we defined $t''=t+\tau_1-\tau_2$ (going from 0 to ∞), and

$$\mathcal{C}_{\substack{(3a)\\(3b)}}=\big[T_+e^{M\tau_2}T_-e^{Mt}e^{-M\tau_2}T_\mp e^{M\tau_1}T_\pm\mathbf{v}^{\mathrm{ss}}\big]_1\,. \tag{25}$$

Integral method at $\tau=0$

We now turn to the zero time delay contribution $S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2)$, which, according to Eq. (21), is given by integrating the correlators (Eqs. (23–25)) over their corresponding domains, changing variables as needed. For instance, the integrals of correlators $\mathcal{C}_{(1i)}$ require the change of variables $t_1\to t$ and $\tau_2\to t'$ (both extending from 0 to ∞). The final expressions for the two integrals (a) and (b) read:

$$\begin{aligned}\mathcal{I}_{\substack{(1a)\\(1b)}}=\frac{\Gamma_1\Gamma_2}{\Gamma_1+\Gamma_2}\frac{1}{(2\pi)^2}\Big[&T_+\frac{-1}{M+(-i\omega_2-\Gamma_1-\frac{\Gamma_2}{2})\mathbf{1}}\\&\times T_\mp\frac{-1}{M+(\pm i\omega_1-i\omega_2-\frac{\Gamma_1+\Gamma_2}{2})\mathbf{1}}\\&\times T_\pm\frac{-1}{M+(-i\omega_2-\frac{\Gamma_2}{2})\mathbf{1}}T_-\mathbf{v}^{\mathrm{ss}}\Big]_1\,.\end{aligned} \tag{26}$$

The second correlators $\mathcal{C}_{(2i)}$ lead to:

$$\begin{aligned}\mathcal{I}_{\substack{(2a)\\(2b)}}=\frac{\Gamma_1\Gamma_2}{\Gamma_1+\Gamma_2}\frac{1}{(2\pi)^2}\Big[&T_+\frac{-1}{M+(-i\omega_2-\Gamma_1-\frac{\Gamma_2}{2})\mathbf{1}}\\&\times T_\mp\frac{-1}{M+(\pm i\omega_1-i\omega_2-\frac{\Gamma_1+\Gamma_2}{2})\mathbf{1}}\\&\times T_-\frac{-1}{M+(\pm i\omega_1-\frac{\Gamma_1}{2})\mathbf{1}}T_\pm\mathbf{v}^{\mathrm{ss}}\Big]_1\,.\end{aligned} \tag{27}$$

And the third correlators $\mathcal{C}_{(3i)}$ lead to:

$$\begin{aligned}\mathcal{I}_{\substack{(3a)\\(3b)}}=\frac{\Gamma_1\Gamma_2}{\Gamma_1+\Gamma_2}\frac{1}{(2\pi)^2}\Big[&T_+\frac{-1}{M+(-i\omega_2-\Gamma_1-\frac{\Gamma_2}{2})\mathbf{1}}\\&\times T_-\frac{-1}{M-\Gamma_1\mathbf{1}}\\&\times T_\mp\frac{-1}{M+(\pm i\omega_1-\frac{\Gamma_1}{2})\mathbf{1}}T_\pm\mathbf{v}^{\mathrm{ss}}\Big]_1\,.\end{aligned} \tag{28}$$

The total correlation function follows from twice the real part of the six previous integrals summed over and exchanging photons:

$$S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2)=2\Re\sum_{i=1,2,3}\Big[\mathcal{I}_{(ia)}+\mathcal{I}_{(ib)}\Big]+[1\leftrightarrow 2]\,. \tag{29}$$

Integral method at $\tau>0$

The finite time-delay contribution $\Delta S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2,\tau)$ requires different domains of integration only for the variables t_2, now ranging from T_1 to T_2, and $t=t_2-t_1$ now ranging from t_2-T_1 to ∞. As a result, the integrals in Eq. (22) depend on τ. The integrals on the correlators $\mathcal{C}_{\substack{(1a)\\(1b)}}$ and $\mathcal{C}_{\substack{(2a)\\(2b)}}$, that we note $\Delta\mathcal{I}_{\substack{(1a)\\(1b)}}$ and $\Delta\mathcal{I}_{\substack{(2a)\\(2b)}}$ give similar results as the corresponding $\mathcal{I}_{\substack{(ia)\\(ib)}}$, but they acquire the τ-dependence in the form of a factor $(\Gamma_1+\Gamma_2)\mathcal{F}(\tau)$, with

$$\mathcal{F}(\tau)=e^{-\Gamma_2\tau}\frac{e^{[M+(-i\omega_2+\frac{\Gamma_2}{2})\mathbf{1}]\tau}-1}{M+(-i\omega_2+\frac{\Gamma_2}{2})\mathbf{1}}\,, \tag{30}$$

that is to be inserted in Eqs. (26), (27) after the first matrix T_+. The integrals on $\mathcal{C}_{\substack{(3a)\\(3b)}}$, on the other hand, are not so straightforward. They are to be separated into two parts: one where $t_2-\tau_2<T_1$, the other

one $t_2 - \tau_2 > T_1$. The first part, with integrals $\int_{T_1}^{T_2} dt_2 \int_{\tau_2}^{\infty} dt \int_{t_2-T_1}^{\infty} d\tau_2 \int_0^{\infty} d\tau_1(\ldots)$, gives rise to a quantity similar to $\Delta\mathcal{I}_{\substack{(ia)\\(ib)}}(\tau)$ with $i = 1, 2$, in that its τ-dependence also consists in the factor $(\Gamma_1 + \Gamma_2)\mathcal{F}(\tau)$ inserted after the first matrix T_+ in Eq. (28). For this reason we note it $\Delta\mathcal{I}_{\substack{(3a)\\(3b)}}(\tau)$. The second part, with integrals $\int_{T_1}^{T_2} dt_2 \int_{t_2-T_1}^{\infty} dt \int_0^{t_2-T_1} d\tau_2 \int_0^{\infty} d\tau_1(\ldots)$, yields two more contributions:

$$\Delta\mathcal{I}_{\substack{(3\alpha)\\(3\beta)}}(\tau) = \frac{\Gamma_1\Gamma_2}{(2\pi)^2}\Big[T_+\mathcal{Z}(\tau) \times \frac{-1}{M - \Gamma_1\mathbf{1}}T_\mp \frac{-1}{M + (\pm i\omega_1 - \frac{\Gamma_1}{2})\mathbf{1}}T_\pm\mathbf{v}^{\mathrm{ss}}\Big]_1, \quad (31)$$

where we introduced the τ-dependent matrix

$$\mathcal{Z}(\tau) = \int_{T_1}^{T_2} dt_2 \int_0^{t_2-T_1} d\tau_2 e^{-\Gamma_2(T_2-t_2)} \times e^{\left(M+(-i\omega_2-\frac{\Gamma_2}{2})\mathbf{1}\right)\tau_2} T_- e^{M(t_2-T_1-\tau_2)}. \quad (32)$$

$\mathcal{Z}(\tau)$ can be calculated for each element $T_-^{kl} \in \{0, 1\}$ of the matrix T_-:

$$\mathcal{Z}_{i,j}(\tau) = e^{-\Gamma_2\tau}\sum_{p,k,l,q}\frac{E_{ip}E^{-1}_{pk}T_-^{kl}E_{lq}E^{-1}_{qj}}{m_p - m_q - i\omega_2 - \frac{\Gamma_2}{2}} \times \left\{\frac{e^{(m_p - i\omega_2 + \frac{\Gamma_2}{2})\tau} - 1}{m_p - i\omega_2 + \frac{\Gamma_2}{2}} - \frac{e^{(m_q+\Gamma_2)\tau} - 1}{m_q + \Gamma_2}\right\}, \quad (33)$$

where E is the matrix of eigenvectors of M, that diagonalises it: $M_{ik} = \sum_p E_{ip}m_pE^{-1}_{pk}$, with m_p the eigenvalues.

Gathering terms with the same τ dependence defines $\Delta\mathcal{I}(\tau) = \sum_{i=1,2,3}\left[\Delta\mathcal{I}_{(ia)}(\tau) + \Delta\mathcal{I}_{(ib)}(\tau)\right]$ which enters in the final result:

$$\Delta S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1; \omega_2, \tau) = 2\Re\Big[\Delta\mathcal{I}(\tau) + \Delta\mathcal{I}_{(3\alpha)}(\tau) + \Delta\mathcal{I}_{(3\beta)}(\tau)\Big]. \quad (34)$$

Sensing method at $\tau = 0$

The intensity correlations between two sensors, $\langle n_1n_2\rangle = \langle\varsigma_1^\dagger\varsigma_1\varsigma_2^\dagger\varsigma_2\rangle$, have the equation of motion:

$$\partial_t\langle n_1n_2\rangle = -(\Gamma_1 + \Gamma_2)\langle n_1n_2\rangle + 2\Re\Big[i\varepsilon_2\langle\varsigma_1^\dagger\varsigma_1\varsigma_2a^\dagger\rangle + i\varepsilon_1\langle\varsigma_1\varsigma_2^\dagger\varsigma_2a^\dagger\rangle\Big]. \quad (35)$$

This leads to the steady state solution:

$$\langle n_1n_2\rangle = \frac{2}{\Gamma_1 + \Gamma_2}\Re\Big[i\varepsilon_2T_+\mathbf{w}[11, 01]\Big]_1 + [1 \leftrightarrow 2]. \quad (36)$$

This solution relies on $\mathbf{w}[11, 01]$ which can be expressed in terms of three lower order correlators:

$$\mathbf{w}[11, 01] = \frac{-1}{M + (-i\omega_2 - \Gamma_1 - \frac{\Gamma_2}{2})\mathbf{1}} \times\Big\{-i\varepsilon_2T_-\mathbf{w}[11, 00] - i\varepsilon_1T_-\mathbf{w}[10, 01] + i\varepsilon_1T_+\mathbf{w}[01, 01]\Big\}, \quad (37)$$

each of which is given by:

$$\mathbf{w}[11, 00] = \frac{-1}{M - \Gamma_1\mathbf{1}} \times\Big\{i\varepsilon_1T_+\mathbf{w}[01, 00] - i\varepsilon_1T_-\mathbf{w}[10, 00]\Big\}, \quad (38)$$

$$\mathbf{w}[10, 01] = \frac{-1}{M + (i\omega_1 - i\omega_2 - \frac{\Gamma_1+\Gamma_2}{2})\mathbf{1}} \times\Big\{-i\varepsilon_2T_-\mathbf{w}[10, 00] + i\varepsilon_1T_+\mathbf{w}[00, 01]\Big\}, \quad (39)$$

and

$$\mathbf{w}[01, 01] = \frac{-1}{M + (-i\omega_1 - i\omega_2 - \frac{\Gamma_1+\Gamma_2}{2})\mathbf{1}} \times\Big\{-i\varepsilon_1T_-\mathbf{w}[00, 01] - i\varepsilon_2T_-\mathbf{w}[01, 00]\Big\}. \quad (40)$$

Finally, we apply a last time the recursive relation Eq. (13) to find the three different correlators involved in the previous expressions:

$$\mathbf{w}[10, 00] = \frac{-1}{M + (i\omega_1 - \frac{\Gamma_1}{2})\mathbf{1}}i\varepsilon_1T_+\mathbf{v}^{\mathrm{ss}}, \quad (41a)$$

$$\mathbf{w}[00, 01] = \frac{-1}{M + (-i\omega_2 - \frac{\Gamma_2}{2})\mathbf{1}}(-i\varepsilon_2T_-)\mathbf{v}^{\mathrm{ss}}, \quad (41b)$$

$$\mathbf{w}[01, 00] = \frac{-1}{M + (-i\omega_1 - \frac{\Gamma_1}{2})\mathbf{1}}(-i\varepsilon_1T_-)\mathbf{v}^{\mathrm{ss}}. \quad (41c)$$

Sensing method at $\tau > 0$

We now consider the case where the second photon is absorbed by sensor 2 with some delay $\tau > 0$ after a first photon is absorbed by sensor 1. The correlator of interest is $\langle n_1(0)n_2(\tau)\rangle$, with equation of motion:

$$\partial_\tau\langle n_1(0)n_2(\tau)\rangle = -\Gamma_2\langle n_1(0)n_2(\tau)\rangle + 2\Re\Big[i\varepsilon_2\langle n_1(0)(\varsigma_2a^\dagger)(\tau)\rangle\Big], \quad (42)$$

with the initial condition in the steady state $\langle n_1(0)n_2(0)\rangle = \langle n_1n_2\rangle$. This solution relies on $\langle n_1(0)(\varsigma_2a^\dagger)(\tau)\rangle$. To compute it, we introduce a vector analogous to Eq. (43) but now consisting of two-time correlators:

$$\mathbf{w}'[11,\mu_2\nu_2](\tau) = \begin{pmatrix} \langle n_1(0)(\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2})(\tau)\rangle \\ \langle n_1(0)(\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}a)(\tau)\rangle \\ \langle n_1(0)(\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}a^\dagger)(\tau)\rangle \\ \langle n_1(0)(\varsigma_2^{\dagger\mu_2}\varsigma_2^{\nu_2}a^\dagger a)(\tau)\rangle \\ \vdots \end{pmatrix}. \quad (43)$$

With this definition, $\langle n_1(0)(\varsigma_2 a^\dagger)(\tau)\rangle$ is the first element of the vector $T_+\mathbf{w}'[11,01](\tau)$. The τ-equation for $\mathbf{w}'[11,01](\tau)$ reads:

$$\partial_\tau \mathbf{w}'[11,01](\tau) = \Big[M + (-i\omega_2 - \frac{\Gamma_2}{2})\mathbf{1}\Big]\mathbf{w}'[11,01](\tau) - i\varepsilon_2 T_- \mathbf{w}'[11,00](\tau)\,, \quad (44)$$

with $\mathbf{w}'[11,00](\tau) = e^{M\tau}\mathbf{w}[11,00]$ with initial condition $\mathbf{w}'[11,01](0) = \mathbf{w}[11,01]$ in the steady state. After some algebra, one arrives to the solution:

$$\mathbf{w}'[11,01](\tau) = e^{\left[M+(-i\omega_2-\frac{\Gamma_2}{2})\mathbf{1}\right]\tau}\mathbf{w}[11,01] - (-i\varepsilon_2)\mathcal{Y}(\tau)\mathbf{w}[11,00]\,, \quad (45)$$

in terms of a matrix $\mathcal{Y}(\tau)$ defined elementwise as:

$$\mathcal{Y}_{ij}(\tau) = \sum_{p,k,l,q} \frac{E_{ip}E^{-1}_{pk}T^{kl}_- E_{lq}E^{-1}_{qj}}{m_p - m_q - i\omega_2 - \frac{\Gamma_2}{2}} \Big\{e^{(m_p - i\omega_2 - \frac{\Gamma_2}{2})\tau} - e^{m_q\tau}\Big\}. \quad (46)$$

Substituting this expression into Eq. (42) and solving it, we obtain:

$$\langle n_1(0)n_2(\tau)\rangle = e^{-\Gamma_2\tau}\langle n_1 n_2\rangle + 2\Re\Big[i\varepsilon_2 T_+\mathcal{F}(\tau)\mathbf{w}[11,01]\Big]_1 + 2\Re\Big[\varepsilon_2^2 T_+\mathcal{Z}(\tau)\mathbf{w}[11,00]\Big]_1\,, \quad (47)$$

where the matrices $\mathcal{F}(\tau)$ and $\mathcal{Z}(\tau)$ are those introduced in the previous section, namely, Eqs. (30) and (33), respectively.

Equality of the integral and sensing methods

We complete the proof by showing that the results from the integration and the sensing methods are the same to leading order in the coupling ε.

First, the case $\tau = 0$. The final expression for $\langle n_1 n_2\rangle$ is obtained by inserting the solutions for the correlators (38–40) into Eq. (36). This leads to the same results as Eq. (29), with the integrals appearing precisely in the following order:

$$\langle n_1 n_2\rangle = \frac{\varepsilon_1^2\varepsilon_2^2}{\Gamma_1\Gamma_2}(2\pi)^2 \times 2\Re\Big\{\mathcal{I}_{(3b)} + \mathcal{I}_{(3a)} + \mathcal{I}_{(2a)} + \mathcal{I}_{(1a)} + \mathcal{I}_{(1b)} + \mathcal{I}_{(2b)}\Big\} + [1 \leftrightarrow 2] = \frac{\varepsilon_1^2\varepsilon_2^2}{\Gamma_1\Gamma_2}(2\pi)^2 S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2)\,. \quad (48)$$

Second, the case $\tau > 0$. For ease of comparison, we rewrite the term $\Delta\mathcal{I}$ in term of the vector $\mathbf{w}[11,01]$ as:

$$\Delta\mathcal{I}(\tau) = \frac{\Gamma_1\Gamma_2}{(2\pi)^2}\Big[T_+\mathcal{F}(\tau)\frac{1}{\varepsilon_1^2(-i\varepsilon_2)}\mathbf{w}[11,01]\Big]_1\,. \quad (49)$$

It is then clear that this expression is equal, up to a constant factor, to the second line in the expression for $\langle n_1(0)n_2(\tau)\rangle$, Eq. (47). Similarly, the term $\Delta\mathcal{I}_{(3\alpha)}(\tau) + \Delta\mathcal{I}_{(3\beta)}(\tau)$ in Eq. (31) can be rewritten as:

$$\Delta\mathcal{I}_{(3\alpha)}(\tau) + \Delta\mathcal{I}_{(3\beta)}(\tau) = \frac{\Gamma_1\Gamma_2}{(2\pi)^2}\Big[T_+\mathcal{Z}(\tau)\frac{1}{\varepsilon_1^2}\mathbf{w}[11,00]\Big]_1\,, \quad (50)$$

and related to the third line in Eq. (47). All together, we can therefore conclude that, to leading order in the couplings:

$$\langle n_1(0)n_2(\tau)\rangle = \frac{\varepsilon_1^2\varepsilon_2^2}{\Gamma_1\Gamma_2}(2\pi)^2 S^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2,\tau)\,. \quad (51)$$

Final remarks

This proof can be generalised to N-photon correlations and/or for finite T_1-time dynamics (instead of a steady state) by repeating these procedures linearly in the number of sensors and integrals. There is no conceptual difference brought by the higher number of variables, but notations become heavy and for the sake of clarity, we have illustrated the proof in the simplest, as well as most relevant cases, of $N = 1$ and 2. Also, nothing in the proof relies on the choice of sensors as two-level systems, which has been made for convenience. As we always examine crossed correlations between them, they could also be, e.g., harmonic oscillators, and provide identical results.

Together with Eqs. (36–41), Eq. (47) provides a semi-analytical result that can be used directly for computations. Although the Hilbert space is not enlarged when using these formulas, they are however awkward to use and set up. Also, the growth in the number of correlators has the same power dependence on the maximum number of excitations allowed in the system than when including the sensors explicitly (it is linear in the Jaynes–Cummings model). The number of correlators increases like 4^N when including N sensors. Benchmarks for $N = 2$ show that the many matrix operations (inversions and multiplications) involved to evaluate the formulas are more costly than solving linear equations as required when including explicitly the sensors. Although this is for a larger set of correlators in the latter case, optimisations such as LU decomposition make sensors a more efficient as well as a conceptually simpler approach. If using the semi-analytical formulas turns out to be more effective in a particular context or for larger N, similar results can be derived for $\langle n_1(0)n_2(\tau_1)\dots n_N(\tau_{N-1})\rangle$ by generalizing Eq. (13) with N sensors to obtain $\mathbf{w}[\mu_1\nu_1,\dots,\mu_N\nu_N]$ recursively.

Finally, the case of N identical sensors reproduces exactly the N-photon correlations, $g^{(N)}$, from a single harmonic sensor (full correlations of the output of a single filter). This can be shown by comparing the presented derivation with N two-level sensors with one where the system is coupled to a single bosonic sensor with associated $\mathbf{w}$ vectors of the type $\mathbf{w}[n,m]$ (where n, $m=0,\dots N$). The results are also seen to be identical to those obtained by substituting $\omega_1=\omega_2=\omega$ and $\Gamma_1=\Gamma_2=\Gamma$ in the formula for $g^{(2)}_{\Gamma_1\Gamma_2}(\omega_1;\omega_2,\tau)$.

FURTHER APPLICATION TO THE JAYNES–CUMMINGS MODEL

The sensing method was illustrated in the text up to three frequencies and for various time delays. Here we provide a supplemental example up to four-photon correlations.

Such high-order correlations are not intuitive to visualise in their most general representation, given that they convey more information and of a much deeper character than single-photon observables. Photons are emitted at all energies and some correlations for particular energies other than $\pm R_n^{\pm}$ are suppressed or, on the contrary, enhanced, meaning that more complicated processes than simple relaxation take place. We reserve to future works the presentation of how such new processes of emission can be identified in the study of frequency resolved correlations, already at the two-photon level, and how these may find new applications to optimise quantum emitters. Here, to keep the discussion succinct, we will focus on the most important processes only, where N photons are detected at precisely the Jaynes–Cummings transitions, that is, we disregard the correlations where one or more photons have an energy which does not correspond to a transition in the ladder.

In Fig. 1, we compare two, three and four-photon correlations of photons with energies corresponding to the possible placements of detectors over all possible transitions. There are 2^{2N-1} configurations, half of them being symmetric with the other half by the interchange of upper and lower polaritons in all rungs. We need only consider, therefore, 2^{2N-2} cases, which are displayed in Fig. 1 representing only the half which detects the upper polariton in the first rung. For instance the leftmost case in panel (c) corresponds to setting four detectors at the energies $\omega_1,\dots,\omega_4$ probing the transitions $|4+\rangle\to|3-\rangle$, $|3+\rangle\to|2-\rangle$, $|2+\rangle\to|1-\rangle$ and $|1+\rangle\to|\mathrm{vac}\rangle$. As in this sequence of detection, polaritons have to swap branch in all rungs, the emission is unlikely and the corresponding coincidence is strongly suppressed.

As discussed in the main text, finite τ are important since correlations may be maximised at nonzero time-delays, when the dynamics of relaxation synchronises with detection. It is however difficult to find the optimising values for $N-1$ independent degrees of freedom when measuring Nth order correlations. We show here that the simplest approximation to fix all delays at zero already leads to useful results which contain the gist of the dynamics. An absolute value of photon correlations has little meaning in itself. It is when compared to other correlations in alternative configurations that a physical meaning can be identified and quantified. Figure 1 shows how, even at equal times, the detection of photons at energies that correspond to a cascade of the Jaynes–Cummings ladder results in giant bunching. These are the points on the right of each panel. On the opposite, as previously described, when the detectors are arranged to click in the sequence that least correspond to a cascade, that is, alternating the type of polariton each time the system goes one rung down, a corresponding giant suppression is obtained. This is an actual antibunching in the case of two-photon detection (a), while with a higher number of photons, the values obtained are larger than one but, again, when compared to the relative values of other transitions, reveal a giant suppression of correlations of over five and ten orders of magnitudes in three and four-photon counting, respectively.

Another remarkable behaviour of these figures is the emergence of a classical behaviour with the increasing number of detected photons, powered by combinatorial growth. While correlations are markedly distinct and varying abruptly in the extreme, low-entropy situations (on both sides of the horizontal axes), the large number of intermediate configurations smoothes out the quantized character and yields a gradual and milder variation as one quantum in the chain of detections is shifted from its precise expected value. The larger the number of photons, the faster is this transition from a discrete, staircase behaviour to a smooth continuous one. These are the transitions shown in black (also with smaller arrows in (c)). These results also reveal that proper sequencing of the detection allows to isolate and magnify its quantum character, even when dealing with a large number of photons. This only hints at the rich physics unravelled by N-photon correlations and at the applications they could bring about.

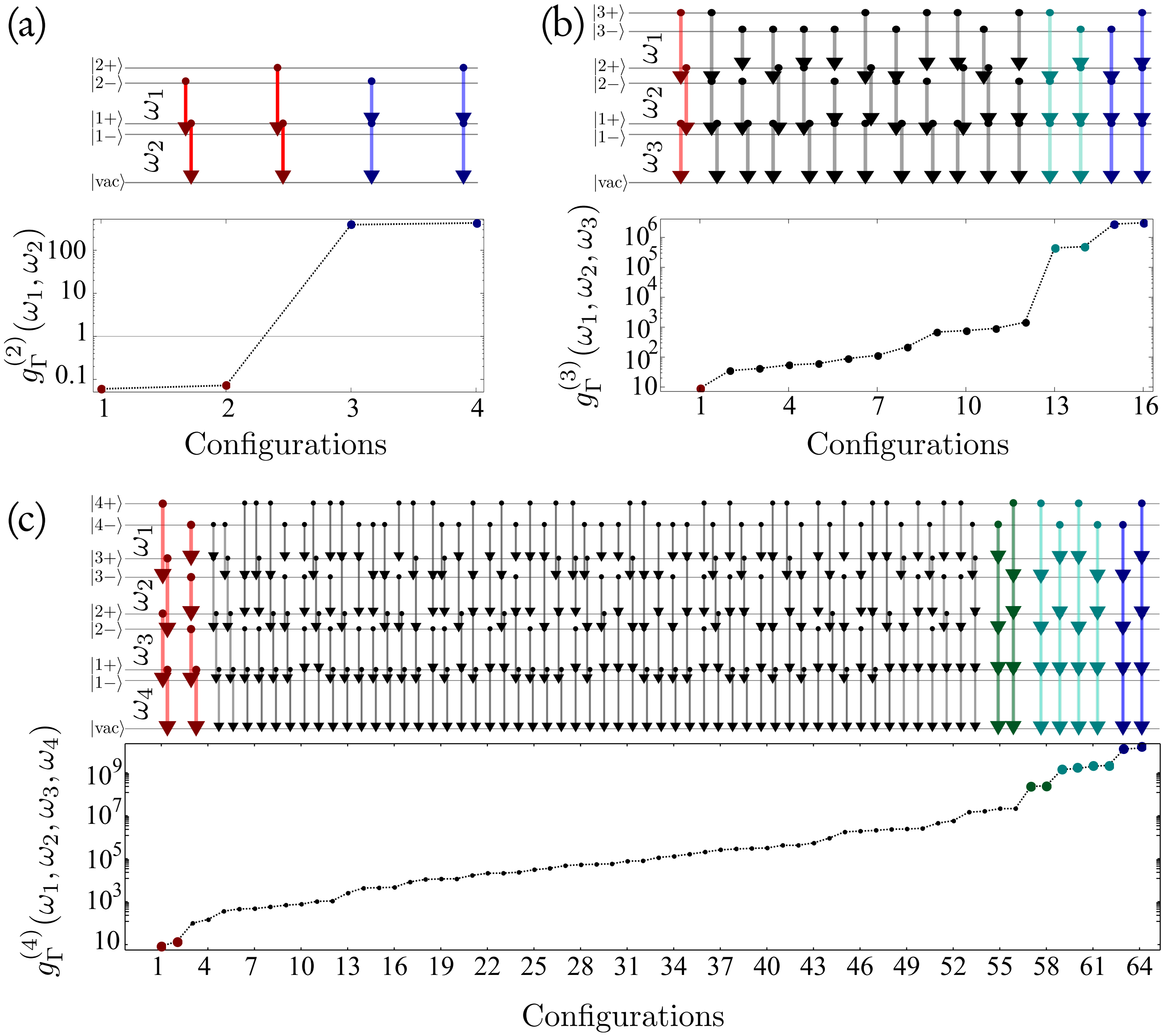

FIG. 1: (Color online) (a) Two-, (b) three- and (c) four-photon correlations in the Jaynes–Cummings model with detection at the frequencies corresponding to transitions in the ladder. For visibility, whenever the arrows would overlap, a small shift has been introduced to assist the eye in tracking the starting and ending points. By probing transitions in a cascade or, on the contrary, from different de-excitation routes, the correlations between the detected photons vary over 3, 5 and 10 orders of magnitudes at the two, three and four-sensor level, respectively. Note that the vertical axes are in log-scale, so even if the variations appear moderate, they are locally important. Only transitions detecting the upper polariton $|1+\rangle \to |\mathrm{vac}\rangle$ in the first rung are shown, those detecting the lower polariton $|1-\rangle \to |\mathrm{vac}\rangle$ are reconstructed by swapping upper and lower polaritons in all rungs. Parameters: $\gamma_a = \gamma_\sigma = 0.001g$, $\Gamma = \gamma_2 = 0.003g$ in the limit of vanishing incoherent pumping of the emitter, $P_\sigma \to 0$.

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