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# Photon entanglement entropy as a probe of many-body correlations and fluctuations

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## ABSTRACT

Recent theories and experiments have explored the use of entangled photons as a spectroscopic probe of physical systems. We describe here a theoretical description for entropy production in the scattering of an entangled biphoton Fock state within an optical cavity. We develop this using perturbation theory by expanding the biphoton scattering matrix in terms of single-photon terms in which we introduce the photon-photon interaction via a complex coupling constant,  $\xi$ . We show that the von Neumann entropy provides a concise measure of this interaction. We then develop a microscopic model and show that in the limit of fast fluctuations, the entanglement entropy vanishes, whereas in the limit of slow fluctuations, the entanglement entropy depends on the magnitude of the fluctuations and reaches a maximum. Our result suggests that experiments measuring biphoton entanglement give microscopic information pertaining to exciton-exciton correlations.

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## I. INTRODUCTION

Experiments using entangled photon pairs as probes of material systems have opened a new arena for both linear and non-linear spectroscopy. Measurements with quantum photons provide sensitive measures of multi-photon processes and many-body correlations.<sup>1–20</sup> This sensitivity can be attributed to the “spooky action at a distance” nature of entangled photons, whereby measurement of one photon gives information about its entangled partner photon through either coincidence detection, interference, or quantum state reconstruction. The fundamental problem we consider is that of an ensemble of two-level atoms in a resonant cavity, interacting with a two photon input Fock state  $|\omega_1, \omega_2\rangle$  to produce a final outgoing photon state that is a superposition of biphoton states of the form

$$|\psi\rangle = \iint d\omega_1 d\omega_2 \mathcal{F}_{out}(\omega_1, \omega_2) |\omega_1, \omega_2\rangle. \quad (1)$$

The amplitude for this process is not simply the product of amplitudes for single photon processes, rather it must reflect the cross correlation between the same field at two different points and at two different times. That is to say, it must consist of both reducible, single photon terms and irreducible cross terms. Our work here is to deduce the general form for this irreducible part and connect it to the microscopic interaction between pairs of atoms in the cavity. Our model draws inspiration from the work by Milonni<sup>21</sup> which considers correlation between two photodetectors, taking into account the time-retardation of the field generated by one atom and felt by the second atom. A simpler version of this which does not take into account the time retardation is presented in Ref. 22 for the case of the direct detection of one photon, given the presence of another and hearkens back to even earlier work by Fermi.<sup>23</sup>

The central focus of this work is the biphoton scattering matrix  $\mathcal{S}^{(2)}(\omega_1, \omega_2; \omega'_1, \omega'_2)$  which transforms a two-photon input

state  $|\omega'_1, \omega'_2\rangle$  into a two-photon output state  $|\omega_1, \omega_2\rangle$  and we develop a formal theory using both frequency-domain and time-domain approaches. Both cases are related to the entanglement entropy of the outgoing biphoton quantum state. Our frequency-domain theory develops from a perturbative expansion in which the single-photon terms are coupled order-by-order via a complex *entanglement parameter*,  $\xi$ , which we take as a measure of the photon-photon coupling mediated by the medium. We also develop a time-domain approach whereby photon-photon entanglement is mediated by cross-correlated spectral fluctuations. We show that in the limit of rapid fluctuations, an entanglement gets destroyed, whereas in the limit of slow fluctuations, the fluctuations produce entangled states with a maximum entropy determined by the spectral width.

## II. TWO-PHOTON SCATTERING AMPLITUDES

The central hypothesis of this paper is that one can directly correlate changes in a biphoton state to many-body interactions within a material system. Let us define an initial biphoton input state to be of the form<sup>34</sup>

$$|\psi_{\text{in}}\rangle = \iint d\omega_1 d\omega_2 \mathcal{F}_{\text{in}}(\omega_1, \omega_2) \hat{B}^\dagger(\omega_1) \hat{B}^\dagger(\omega_2) |0\rangle. \quad (2)$$

Here, we define  $\mathcal{F}_{\text{in}}$  as the biphoton amplitude and as such  $|\psi_{\text{in}}\rangle$  is a superposition of Fock states. This state interacts with a sample to produce a biphoton output state

$$|\psi_{\text{out}}\rangle = \iint d\omega'_1 d\omega'_2 \left[ \iint d\omega_1 d\omega_2 \mathcal{S}^{(2)}(\omega_1, \omega_2; \omega'_1, \omega'_2) \mathcal{F}_{\text{in}}(\omega'_1, \omega'_2) \right] \hat{B}^\dagger(\omega'_1) \hat{B}^\dagger(\omega'_2) |0\rangle. \quad (3)$$

Here, we denote photon creation operators  $\hat{B}^\dagger(\omega_1)$  and  $\hat{B}^\dagger(\omega_2)$  acting on the photon vacuum state  $|0\rangle$ . We shall leave the exact representation of the scattering matrix,  $\mathcal{S}^{(2)}(\omega_1, \omega_2; \omega'_1, \omega'_2)$ , undefined at the moment.

The key to understanding entanglement is separability. An entangled two-particle state cannot be written as a product of two single-particle states. This separability can be quantified by performing Singular Value Decomposition (SVD) on the two-particle amplitude function. This is also termed Schmidt decomposition. Suppose we write the input amplitude as  $\mathcal{F}(\omega_1, \omega_2)$  and perform SVD on this function

$$\mathcal{F}(\omega_1, \omega_2) = \sum_n r_n U_n(\omega_1) V_n(\omega_2), \quad (4)$$

where  $U_n$  and  $V_n$  are the SVD left and right eigenvectors and the  $r_n$  expansion coefficients are the SVD eigenvalues. These we assume to be normalized to unity without loss of generality. The von Neumann entropy provides a measure of the entanglement

$$S = - \sum_n r_n^2 \ln r_n^2. \quad (5)$$

If only one term contributes, the function is separable and  $S = 0$ . Otherwise, if multiple terms contribute to the SVD expansion, the system is entangled and the resulting  $S > 0$ . Now suppose we allow the biphoton state to interact with the sample but such that each interaction is independent of the other interaction. The resulting outgoing photon state must be of the form

$$\mathcal{F}'(\omega_1, \omega_2) = \int d\omega'_1 \int d\omega'_2 G(\omega_1, \omega'_1) H(\omega_2, \omega'_2) \mathcal{F}(\omega'_1, \omega'_2). \quad (6)$$

Again, performing SVD on this new function,

$$\mathcal{F}'(\omega_1, \omega_2) = \int d\omega'_1 \int d\omega'_2 G(\omega_1, \omega'_1) H(\omega_2, \omega'_2) \times \sum_k r_k U_k(\omega'_1) V_k(\omega'_2). \quad (7)$$

The function remains separable since we can rearrange this as

$$\mathcal{F}'(\omega_1, \omega_2) = \sum_k r_k \left( \int d\omega'_1 G(\omega_1, \omega'_1) U_k(\omega'_1) \right) \times \left( \int d\omega'_2 H(\omega_2, \omega'_2) V_k(\omega'_2) \right). \quad (8)$$

Moreover, SVD returns the same set of coefficients although the SVD basis vectors are different. The entanglement does not change because there is no cross talk between the different degrees of freedom. If we introduce any cross correlations between the degrees of freedoms, there is a resultant change in entanglement entropy. For example, if we were to write the biphoton amplitude in the form of the products of single-photon amplitudes

$$\mathcal{F}'(\omega_1, \omega_2) = \int d\omega'_1 \int d\omega'_2 \mathcal{S}^{(1)}(\omega_1, \omega'_1) \mathcal{S}^{(1)}(\omega_2, \omega'_2) \mathcal{K}(\omega_1, \omega'_1; \omega_2, \omega'_2). \quad (9)$$

It is clear that the SVD of this new amplitude will produce a very different outgoing state in which any change in the entropy must be induced by cross correlations within the system. Experimentally, by measuring the biphoton entropy with and without a sample present will produce a measure of the magnitude and nature of such microscopic interactions. This is, of course, a central goal of spectroscopic investigation of physical systems. We now develop connections between the scattered biphoton amplitude  $\mathcal{S}^{(2)}$  and the spectral response of the system.

### A. Diagrammatic expansion of the two-photon scattering matrix

In general, we can write the elastic scattering of a single photon through a resonant medium with a Lorentzian line shape in the form<sup>10</sup>

$$\mathcal{S}^{(1)}(\omega, \omega') = \exp(\mathcal{A}(\omega)) \delta(\omega - \omega'), \quad (10)$$

where

$$\mathcal{A}(\omega) = - \frac{ib}{(\omega_o - \omega) + iy} \quad (11)$$

is the Fourier transform of the free induction decay

$$\mathcal{A}(t) = -\sqrt{2\pi} b e^{-\gamma t} e^{i\omega_o t} \quad (12)$$

for  $t > 0$  of an oscillator with frequency  $\omega_o$  and dephasing rate  $\gamma$ .  $b = \alpha L \gamma / 2$ , where  $\alpha L$  is the optical thickness and  $\alpha$  is a Bouguer coefficient. Consequently, if two independent (unentangled) photons are scattered from the resonant medium, we anticipate a scattering amplitude of

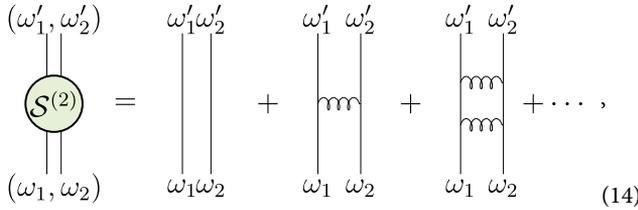
$$\mathcal{S}^{(2)}(\omega_1, \omega_2; \omega'_1, \omega'_2) = \mathcal{S}^{(1)}(\omega_1) \mathcal{S}^{(1)}(\omega_2) \delta(\omega_1 - \omega'_1) \delta(\omega_2 - \omega'_2). \quad (13)$$

In this case, two independent photons are transmitted without any interaction leading to them being unentangled.

Suppose, however, that interactions leading to entanglement are weak such that we can write the two photon scattering amplitude as perturbation expansion of the form

$$S^{(2)} = S_0^{(2)} + S_0^{(2)} \mathcal{V} S_0^{(2)} = S_0^{(2)} + S_0^{(2)} \mathcal{V} S_0^{(2)} + S_0^{(2)} \mathcal{V} S_0^{(2)} \mathcal{V} S_0^{(2)} + \dots,$$

whereby  $S_0^{(2)} = S^{(1)} S^{(1)}$  is separable into single photon terms and  $\mathcal{V}$  mediates the interaction between photon pairs via the resonant cavity. This suggests the following diagrammatic expansion:



$$(14)$$

where solid lines are  $S^{(1)}$  free propagators and springs denote the interaction. Such interactions introduce nonlinearity in the form of cross correlations (i.e., scattering) between otherwise uncoupled single photon processes.

For the special case of elastic scattering,  $\mathcal{V}(\omega)$  contributes a phase-shift of the form

$$\mathcal{V}(\omega) = \xi e^{i\phi} \delta(\omega) \quad (15)$$

but does not create a frequency shift. Then only the term at  $\omega = 0$  will contribute (so that  $\omega_1 = \omega'_1$  and  $\omega_2 = \omega'_2$ )

$$S_1^{(2)} = \xi e^{i\phi} S^{(1)}(\omega_1) S^{(1)}(\omega_2) S^{(1)}(\omega_1) S^{(1)}(\omega_2). \quad (16)$$

Iterating this,

$$S_n^{(2)} = \mathcal{V}^n (S_0^{(2)})^{n+1} = \xi^n e^{in\phi} (S^{(1)}(\omega_1))^{n+1} (S^{(1)}(\omega_2))^{n+1}. \quad (17)$$

Taking

$$S^{(1)}(\omega_i) = \exp\left[-\frac{ib}{(\omega_o - \omega_i) + i\gamma}\right] = z_i \quad (18)$$

is a complex number that is determined by the input photon frequency. Thus, the whole perturbation series becomes

$$S^{(2)} = z_1 z_2 \sum_{n=0}^{\infty} (\xi e^{i\phi} z_1 z_2)^n. \quad (19)$$

Setting  $q = \xi e^{i\phi} z_1 z_2$  and assuming  $|q| < 1$ , then the series can be summed exactly

$$S^{(2)} = \frac{z_1 z_2}{1 - q}. \quad (20)$$

Writing this in terms of the  $S^{(1)}$  functions, we obtain

$$S^{(2)}(\omega_1, \omega_2; \omega_1, \omega_2) = \frac{S^{(1)}(\omega_1) S^{(1)}(\omega_2)}{1 - \xi S^{(1)}(\omega_1) S^{(1)}(\omega_2) e^{i\phi}}. \quad (21)$$

We shall refer to  $\xi$  as the *entanglement parameter*. When  $\xi = 0$ ,

$$S^{(2)}(\omega_1, \omega_2) = \exp[\mathcal{A}^{(1)}(\omega_1)] \exp[\mathcal{A}^{(1)}(\omega_2)] = S^{(1)}(\omega_1) S^{(1)}(\omega_2) \quad (22)$$

is separable in terms of the individual photon amplitudes. It is worth mentioning that the output biphoton state is not normalized due to the entanglement parameter  $\xi$ . Therefore, the proposed formalism could account for signal attenuation and disentanglement effect resulting from the matter-mediated photon coupling.

The expansion in Eq. (19) is in the form of a Schmidt decomposition, which we shall discuss in Sec. II B 2. Taking  $\phi \rightarrow 0$  for convenience and letting  $\xi < 1$ , it becomes trivial to write that the normalized Schmidt coefficients are given by as  $\lambda_n^2 = \xi^{2n} (1 - \xi^2)$  producing a von Neumann entropy

$$S = -\left(\frac{\xi^2 \ln \xi^2}{1 - \xi^2} + \ln(1 - \xi^2)\right). \quad (23)$$

This shows that there exists a 1:1 correspondence between the nonlinear coupling  $\xi$  and the resultant von Neumann entropy,  $S$ .

In principle, the phase  $\phi$  introduced in Eq. (15) depends upon the microscopic details of the system, such as the relative orientation of the atomic or molecular scattering sites within the sample, and may merely be a random quantity. Averaging over phase, we write

$$\overline{S^{(2)}}(\omega_1, \omega_2; \omega_1, \omega_2) = \left\langle \frac{S^{(1)}(\omega_1) S^{(1)}(\omega_2)}{1 - \xi S^{(1)}(\omega_1) S^{(1)}(\omega_2) e^{i\phi}} \right\rangle. \quad (24)$$

Writing this again as a geometric series,

$$\overline{S^{(2)}}(\omega_1, \omega_2; \omega_1, \omega_2) = S^{(1)}(\omega_1) S^{(1)}(\omega_2) \times \sum_{n=0}^{\infty} \langle \xi^n S^{(1)}(\omega_1)^n S^{(1)}(\omega_2)^n e^{in\phi} \rangle. \quad (25)$$

Suppose that the phase  $\phi$  is uniform over  $[0, 2\pi)$ , then

$$\langle e^{in\phi} \rangle = \int_0^{2\pi} \frac{e^{in\phi}}{2\pi} d\phi = \delta_{n0}.$$

In this case, the relative phase is completely randomized and the biphoton amplitude collapses *exactly* into the product of two single photon terms

$$\overline{S^{(2)}}(\omega_1, \omega_2; \omega_1, \omega_2) \rightarrow S^{(1)}(\omega_1) S^{(1)}(\omega_2). \quad (26)$$

On the other hand, suppose the phase is normally distributed about a central value, which we can take to be zero, i.e.,  $\overline{\phi} = 0$  and  $\overline{\phi^2} = \sigma^2$ . Here, the average over  $\phi$  can be cast as

$$\langle e^{in\phi} \rangle = \left(1 - \frac{n^2 \overline{\phi^2}}{2!} + \frac{n^4 \overline{\phi^4}}{4!} - \dots\right).$$

Writing this in terms of the second moment,

$$\begin{aligned} \langle e^{in\phi} \rangle &= \sum_{k=0}^{\infty} (-1)^k (n\sigma)^{2k} \frac{(2k-1)!!}{(2k)!} \\ &= \sum_{k=0}^{\infty} (-1)^k \frac{(n\sigma)^{2k}}{2^k k!} = e^{-(n\sigma)^2/2}. \end{aligned} \quad (27)$$

This gives

$$\bar{\mathcal{S}}^{(2)}(\omega_1, \omega_2; \omega_1, \omega_2) = \mathcal{S}^{(1)}(\omega_1)\mathcal{S}^{(1)}(\omega_2)\mathcal{K}^{(2)}(\omega_1, \omega_2; \omega_1, \omega_2), \quad (28)$$

where

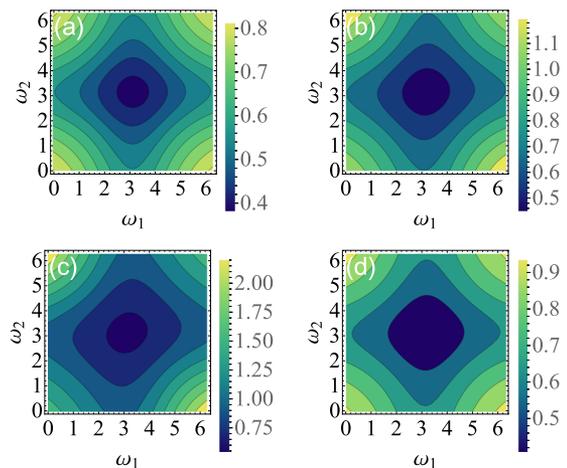
$$\mathcal{K}^{(2)}(\omega_1, \omega_2; \omega_1, \omega_2) = \sum_{n=0}^{\infty} \xi^n e^{-n^2\sigma^2/2} (\mathcal{S}^{(1)}(\omega_1)\mathcal{S}^{(1)}(\omega_2))^n \quad (29)$$

denotes a scattering kernel that reflects the response of the media due to the mode cross correlations facilitating photon-photon scattering and for all intents and purposes is responsible for entanglement.

Plots for this are shown in Fig. 1 for the case of a model system with spectral parameters for Eq. (18):  $\omega_o = \pi$ ,  $b = 1$ , and  $\gamma = 2$ . In each case, we assume that an input of the two-photon Fock state with amplitude  $\mathcal{F}_{\text{in}}(\omega_1, \omega_2)$  is transformed to an output biphoton state with the amplitude up to the factor of  $\mathcal{S}^{(2)}$ , which is correlated along  $\omega_1 = \omega_2$ . For the case of Gaussian noise, the final state is correlated in frequency, as shown in Fig. 1 for various choices of spectral parameters. Furthermore, one can conclude that the resulting state is not necessarily separable into the product of two functions due to the nontrivial von Neumann entropy  $S$  (cf. Sec. II B 2), which gives a useful means of quantifying the entanglement of these states. Schmidt decomposition of Eq. (28) gives Fig. 2 where we have plotted the von Neumann entropy with respect to the interaction parameter  $\xi$ , the Gaussian noise  $\sigma$ , and the dephasing factor  $\gamma$  involved in single photon amplitude  $\mathcal{S}^{(1)}$ . Generally, increasing  $\xi$  leads to an increase in entanglement for a given amount of noise  $\sigma$ , whereas shorter dephasing time  $1/\gamma$  leads to disentanglement, as shown in Fig. 2. An expression for the von Neumann entropy for this case is presented in Appendix A.

## B. Second cumulant model for two-photon scattering amplitude

We now dig deeper and develop a fully microscopic model for entropy production in biphoton scattering. In the context of a previous work,<sup>8</sup> we now consider the case of an ensemble of two-level atoms within a pumped optical microcavity probed by a 2-photon Fock state scattered forward by the cavity. In the time-domain, the input state on the left boundary of the cavity can be represented



**FIG. 1.** Absolute values of two-photon scattering functions  $\mathcal{S}^{(2)}$  for squeezed biphotons with Gaussian noise interactions: (a)  $\xi = 0.1$ ,  $\sigma = 0.1$ ; (b)  $\xi = 0.5$ ,  $\sigma = 0.1$ ; (c)  $\xi = 0.9$ ,  $\sigma = 0.1$ ; and (d)  $\xi = 0.5$ ,  $\sigma = 1.0$ . In each case, we take parameters for  $\mathcal{S}^{(1)}$  in Eq. (18):  $\omega_o = \pi$ ,  $b = 1$ , and  $\gamma = 2$ .

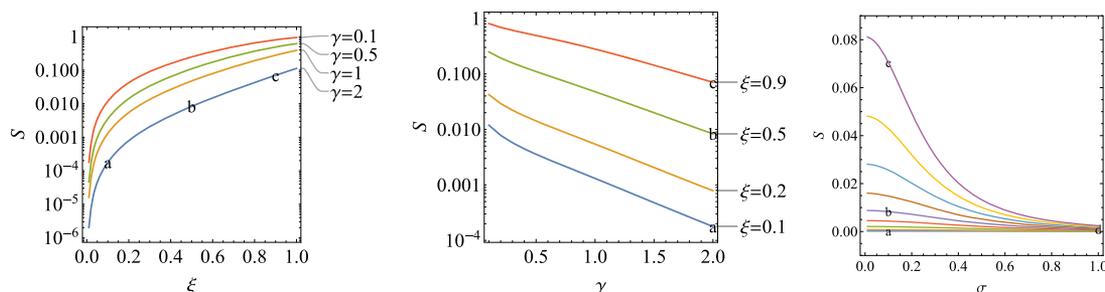
as

$$|\psi_{\text{in}}\rangle = \iint dt_1 dt_2 \mathcal{F}(t_1, t_2) \hat{b}_{1,\text{in}}^\dagger(t_1) \hat{b}_{2,\text{in}}^\dagger(t_2) |0\rangle, \quad (30)$$

where the cavity input photon operator<sup>8</sup>

$$\hat{b}_{j,\text{in}}^\dagger(t) = - \int \frac{d\omega_j}{\sqrt{2\pi}} \hat{B}^\dagger(\omega_j) e^{i\omega_j t} \quad (31)$$

is the Fourier transform of the external photon mode operator  $\hat{B}_j^\dagger(\omega)$  entering Eq. (2). It is noteworthy that substitution of Eq. (31) into Eq. (30) and subsequent integration over the time variables exactly results in the frequency domain representation given by Eq. (2) where  $\mathcal{F}(\omega_1, \omega_2)$  is identified as the Fourier transform of  $\mathcal{F}(t_1, t_2)$ . Following the input output formalism,<sup>25,26</sup> we identify the boundary condition on the left boundary of the cavity with the mode leakage rate  $\kappa_j$  as  $\hat{b}_{j,\text{in}}^\dagger(t) + \hat{b}_{j,r}^\dagger(t) = \sqrt{\kappa_j} \hat{b}_j^\dagger(t)$ . This connects the input photon operator defined above, the reflected photon operator  $\hat{b}_{j,r}^\dagger(t)$ , and the cavity photon operator  $\hat{b}_j^\dagger(t)$ . The



**FIG. 2.** Entanglement entropy  $S_{\Psi}$  [Eq. (57)] vs coupling  $\xi$  (left) for the case of Gaussian noise, dephasing factor  $\gamma$  (middle), and noise strength  $\sigma$  (right). Points a–d correspond to specific biphoton states shown in Figure 1.

reflected mode is not measured in experiment and will not be considered below. However, the cavity mode operator contains information on the interactions within the cavity and will be determined below.

Time domain cavity output operators can be expressed in terms of the photon operators outside the cavity as<sup>8</sup>

$$\hat{b}_{j,\text{out}}^\dagger(t) = \int \frac{d\omega_j}{\sqrt{2\pi}} \hat{B}^\dagger(\omega_j) e^{i\omega_j(t-t_j)}, \quad (32)$$

representing free photon modes  $\hat{B}^\dagger(\omega_j)$  outside the cavity propagated back in time from  $t_j$  to actual measurement time  $t$ . The output state introduced in the frequency domain as

$$|\Psi_{\text{out}}\rangle = \iint d\omega'_1 d\omega'_2 \mathcal{F}_{\text{out}}(\omega'_1, \omega'_2) \hat{B}^\dagger(\omega'_1) \hat{B}^\dagger(\omega'_2) |0\rangle \quad (33)$$

can be represented in terms of the adopted time-domain operators using Eq. (32). This gives

$$|\Psi_{\text{out}}\rangle = \iint d\omega_1 d\omega_2 \iint dt_1 dt_2 \mathcal{F}_{\text{out}}(t'_1, t'_2) e^{-i\omega_1(t_1-t_j)} \times e^{-i\omega_2(t_2-t_j)} \langle \hat{b}_{1,\text{out}}^\dagger(t_1) \hat{b}_{2,\text{out}}^\dagger(t_2) \rangle |0\rangle, \quad (34)$$

where angle brackets describe the average of the output operators over the material induced cavity mode fluctuations. Similar to the input mode, we establish the following boundary condition on the cavity right boundary:  $\hat{b}_{j,\text{out}}^\dagger(t) = \sqrt{\kappa_j} \hat{b}_j^\dagger(t)$ . This condition assumes the same cavity leakage,  $\kappa_j$ , and no input photons on the right. Taking into account this boundary condition, evaluation of the output state using Eq. (34) reduces to the evaluation of the time domain correlation function of the cavity mode operators that will directly result in the evaluation of the desired scattering amplitude.

We adopt the following stochastic Hamiltonian to describe cavity photon modes  $\hat{b}_j^\dagger$  coupled to the input and output biphoton states:

$$\hat{H}_{\text{ph}} = \hbar \sum_{j=1,2} (\omega_j + \delta\hat{\omega}_j) \hat{b}_j^\dagger \hat{b}_j. \quad (35)$$

Here,  $\delta\hat{\omega}_j = \delta\hat{\omega}_j(t)$  is the time-dependent photon frequency fluctuations of each mode. Appendix B provides an example connecting such a generic Hamiltonian with a microscopic Hamiltonian describing photon wave packet scattering by fluctuations of delocalized polariton modes within the cavity. For the sake of generality, we defined the polariton mode fluctuations as operators, which requires handling of  $\delta\hat{\omega}_j = \delta\hat{\omega}_j(t)$  as operators. The  $c$ -number stochastic limit is trivial.

Applying the input-output formalism to the cavity modes described by the Hamiltonian (35), one obtains the quantum Langevin equation<sup>8,27</sup>

$$\frac{\partial}{\partial t} \hat{b}_j^\dagger(t) = i(\tilde{\omega}_j + \delta\hat{\omega}_j) \hat{b}_j^\dagger(t) + \sqrt{\kappa_j} \hat{b}_{j,\text{in}}^\dagger(t), \quad (36)$$

with  $\tilde{\omega}_j = \omega_j + i\kappa_j/2$ . Assuming that the fluctuation dynamics occurs with the rate much faster than the cavity leakage, one can formally integrate Eq. (36) resulting in

$$\hat{b}_{j,\text{out}}^\dagger(t) = \sqrt{\kappa_j} \int_0^t dt' e^{i\tilde{\omega}_j(t-t')} \exp_+ \left[ i \int_{t'}^t d\tau \delta\hat{\omega}_j(\tau) \right] \hat{b}_{j,\text{in}}^\dagger(t'), \quad (37)$$

where  $\exp_+[\dots]$  is a positive time ordered exponential. Here, the cavity right boundary condition is also applied to express the cavity mode operator in terms of the cavity output mode.

According to Eq. (37), the output single and two-photon operators averaged over the fluctuations can be represented as

$$\langle \hat{b}_{j,\text{out}}^\dagger(t) \rangle = \int_0^\infty dt' \mathcal{S}_j^{(1)}(t, t') f_j(t') \hat{b}_{j,\text{in}}^\dagger(t'), \quad (38)$$

$$\langle \hat{b}_{1,\text{out}}^\dagger(t_1) \hat{b}_{2,\text{out}}^\dagger(t_2) \rangle = \int_0^\infty dt'_1 \int_0^\infty dt'_2 \mathcal{S}^{(2)}(t_1 t_2, t'_1 t'_2) \mathcal{F}(t'_1, t'_2) \hat{b}_{1,\text{in}}^\dagger(t'_1) \hat{b}_{2,\text{in}}^\dagger(t'_2), \quad (39)$$

where we assume that  $\mathcal{F}(\omega_1, \omega_2) = f_1(\omega_1) f_2(\omega_2)$  for an initially unentangled photon pair. The single- and two-photon scattering amplitudes entering Eqs. (38) and (39), respectively, read

$$\mathcal{S}_j^{(1)}(t, t') = \theta(t - t') e^{i\tilde{\omega}_j(t-t')} \left\langle \exp_+ \left[ i \int_{t'}^t d\tau \delta\hat{\omega}_j(\tau) \right] \right\rangle, \quad (40)$$

$$\mathcal{S}^{(2)}(t_1 t_2, t'_1 t'_2) = \theta(t_1 - t'_1) \theta(t_2 - t'_2) e^{i\tilde{\omega}_1(t_1-t'_1) + i\tilde{\omega}_2(t_2-t'_2)} \times \left\langle \exp_+ \left[ i \int_{t'_1}^{t_1} d\tau \delta\hat{\omega}_1(\tau) \right] \exp_+ \left[ i \int_{t'_2}^{t_2} d\tau \delta\hat{\omega}_2(\tau) \right] \right\rangle. \quad (41)$$

Here,  $\theta(t)$  is the Heaviside theta-function and angle brackets indicate average over the frequency fluctuations. Substitution of Eqs. (39) and (41) into Eq. (34) provides an expression for the output biphoton state in terms of the scattering amplitude and biphoton input states.

Diagrammatic techniques can be developed for single- and two-photon scattering amplitudes via power series expansion of the exponentials in Eqs. (40) and (41). Instead, we adopt a second cumulant approximation setting all odd point correlation functions in the expansion to zero and partitioning the rest into various products of two-point correlation functions. Summation of the resulting power series gives rise to the following representation of the single- and two-photon scattering amplitudes:<sup>28,29</sup>

$$\mathcal{S}_j^{(1)}(t, t') = e^{i\tilde{\omega}_j(t-t') - g_j(t, t')}, \quad (42)$$

$$\mathcal{S}^{(2)}(t_1 t_2, t'_1 t'_2) = e^{i\tilde{\omega}_1(t_1-t'_1) + i\tilde{\omega}_2(t_2-t'_2)} \times e^{-g_1(t_1, t'_1) - g_2(t_2, t'_2) - g_{12}(t_1 t_2, t'_1 t'_2)}, \quad (43)$$

respectively. Accordingly, the two-photon scattering amplitude can be factorized as

$$\mathcal{S}^{(2)}(t_1 t_2, t'_1 t'_2) = \mathcal{S}_1^{(1)}(t_1, t'_1) \mathcal{S}_2^{(1)}(t_2, t'_2) \mathcal{K}^{(2)}(t_1 t_2, t'_1 t'_2), \quad (44)$$

where the time-domain scattering kernel

$$\mathcal{K}^{(2)}(t_1 t_2, t'_1 t'_2) = e^{-g_{12}(t_1 t_2, t'_1 t'_2)} \quad (45)$$

is introduced.

In Eqs. (42) and (43), the second cumulant function  $g_j$  depends on the  $j$ th frequency autocorrelation function as

$$g_j(t, t') = \int_{t'}^t d\tau_1 \int_{t'}^{\tau_1} d\tau_2 \langle \delta\hat{\omega}_j(\tau_1) \delta\hat{\omega}_j(\tau_2) \rangle. \quad (46)$$

Noteworthy, the integration over  $d\tau_1$  and  $d\tau_2$  is time-ordered insuring causality for a single-photon propagation. The second cumulant function entering the scattering kernel [Eq. (43)] is

$$g_{12}(t_1 t_2, t'_1 t'_2) = \int_{t'_1}^{t_1} d\tau_1 \int_{t'_2}^{t_2} d\tau_2 \langle \delta\hat{\omega}_1(\tau_1) \delta\hat{\omega}_2(\tau_2) \rangle. \quad (47)$$

This accounts for the cross correlations between different photon modes and as we show below affects the photon pair entanglement. In contrast to Eq. (46), here integration over  $d\tau_1$  and  $d\tau_2$  lacks time ordering indicating that the photon cross correlations are not casual. Notice that the four-point time dependence of the second cumulant indicates that the scattering kernel is responsible for a *nonlinear* media response that mixes four electric field amplitudes. If the cross correlation function is zero,  $\mathcal{K}_{12} = 1$  and the two-photon scattering amplitude factorizes to a product of two linear, i.e., single-photon, scattering amplitudes.

### 1. Stochastic Kubo-Anderson model

For further analysis, we adopt the Kubo-Anderson stochastic model which is often used in spectroscopic line shape analysis.<sup>28</sup> This model treats fluctuations as commuting random variables, whose time evolution is a Gaussian stochastic process, thereby making a second cumulant expansion exact. Following this approach, we set

$$\langle \delta\omega_j(\tau_1) \delta\omega_j(\tau_2) \rangle = \sigma_j^2 e^{-|\tau_1 - \tau_2|/\bar{\tau}_j}, \quad (48)$$

$$\langle \delta\omega_1(\tau_1) \delta\omega_2(\tau_2) \rangle = \sigma_{12}^2 e^{-|\tau_1 - \tau_2|/\bar{\tau}_{12}}, \quad (49)$$

where  $\sigma_j^2 = \langle \delta\omega_j^2(0) \rangle$  and  $\sigma_{12}^2 = \langle \delta\omega_1(0) \delta\omega_2(0) \rangle$  ( $\bar{\tau}_j$  and  $\bar{\tau}_{12}$ ) being single-mode and cross-mode variances (correlation times), respectively. The representation of the single photon amplitudes is not essential for the analysis below, and the details of the derivation of the second cumulant functions in Eqs. (46) and (47) for the correlation functions given in Eqs. (48) and (49) are provided in Appendix C. Next, we discuss the limiting cases.

In the limit of fast modulation where  $\sigma_{12} \bar{\tau}_{12} \ll 1$ , only the following four time-ordered contributions (denoted by  $g_{12}^\pm$ ) survive:

$$e^{-g_{12}^+(t_1 t_2 t'_1 t'_2)} = e^{-\gamma_{12}(t_2 - t'_1)}, \quad (50)$$

$$e^{-g_{12}^+(t_1 t_2 t'_2 t'_1)} = e^{-\gamma_{12}(t_2 - t'_2)}, \quad (51)$$

$$e^{-g_{12}^+(t_2 t_1 t'_2 t'_1)} = e^{-\gamma_{12}(t_1 - t'_2)}, \quad (52)$$

$$e^{-g_{12}^+(t_2 t_1 t'_1 t'_2)} = e^{-\gamma_{12}(t_1 - t'_1)}. \quad (53)$$

In the limit of slow modulation for the mode cross correlation, i.e.,  $\sigma_{12} \bar{\tau}_{12} \gg 1$ , only terms that are quadratic in time contribute to the second cumulant function. In this case, the scattering kernel of the two-photon scattering amplitude acquires a Gaussian form

$$\mathcal{K}^{(2)}(t_1 t_2, t'_1 t'_2) = e^{-\sigma_{12}^2 (t_1 - t'_1)(t_2 - t'_2)} \quad (54)$$

in which the decay is determined by the cross correlation  $\sigma_{12}^2 = \langle \delta\omega_1(0) \delta\omega_2(0) \rangle$  magnitude.

### 2. Entanglement entropy analysis

Using the scattering kernel of two-photon scattering amplitude introduced in Eq. (45), we can compute the von Neumann entropy for the scattered biphoton state, whereas above, we computed this in the frequency domain, and the scattering matrix  $S$  is invariant under unitary transformations, including the Fourier transform. This implies that we are free to evaluate the entropy directly from the time-correlation functions. This can be accomplished by performing a Schmidt decomposition of the scattering amplitude in Eq. (44) into separable components. Since this is a product of separable and non-separable terms, we only need to decompose the scattering kernel [Eq. (45)] involving  $g_{12}$ ,

$$e^{-g_{12}(t_1, t_2, t'_1, t'_2)} = \sum_k r_k \phi_k(t_1 - t'_1) \psi_k(t_2 - t'_2), \quad (55)$$

where the function sets  $\{\phi_k(t_1 - t'_1)\}$  and  $\{\psi_k(t_2 - t'_2)\}$  form an orthonormal basis of Schmidt modes and  $\{r_k\}$  is a set of the mode weights. The mode weights provide a useful way to quantify the entanglement between photons. If we write  $\lambda_k = r_k/\sqrt{\Omega}$  as the set of normalized Schmidt coefficients such that

$$\sum_k \lambda_k^2 = 1, \quad (56)$$

we can write the von Neumann entropy as

$$S = - \sum_k \lambda_k^2 \ln(\lambda_k^2). \quad (57)$$

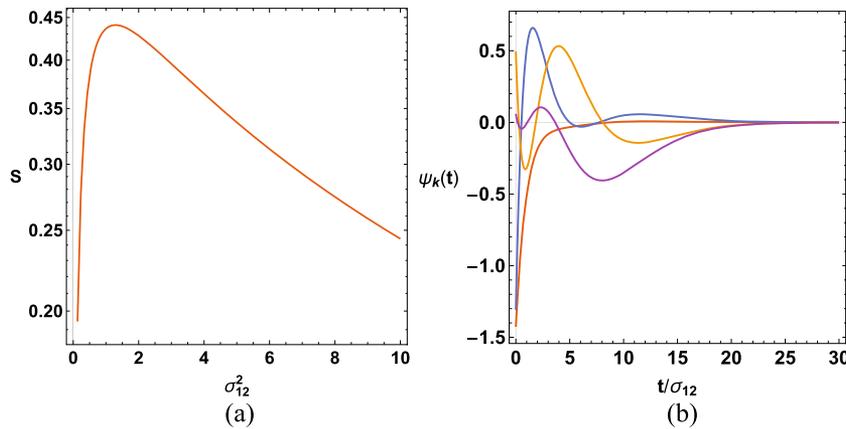
If the state is *separable*, then the entropy is exactly zero and one and only one of the  $\lambda_k^2 = 1$ , and the rest are exactly equal to zero. Moreover,  $S = \ln N$ , where  $N$  is the dimensionality of the Hilbert-space spanned by the basis functions. In other words, increasing  $S$  implies that more and more pairs of Schmidt basis functions are needed to reconstruct the original function.

In the limit of fast modulation, where  $\sigma_{12} \bar{\tau}_{12} \ll 1$ , the exponent of the cross correlation function is separable in terms of the times [Eqs. (50)–(53)], and consequently, the entropy of the outgoing state is exactly equal to 0. This makes sense since in this limit the cross correlation function depends only upon the intermediate two times in the time-ordering. In other words, the only way for photon 1 to interact with photon 2 is if the polarization created by the first persists long enough to influence the second photon. Else, no entanglement can be produced.

In the limit of slow modulation, the cross correlation depends upon all 4 times [Eq. (54)] and cannot be separable into a pair of functions involving only  $t_1 - t'_1$  and  $t_2 - t'_2$ . Here, we first expand Eq. (54) as a sum product of Laguerre polynomials taking  $x = t_1 - t'_1$  and  $y = t_2 - t'_2$ ,

$$e^{-\sigma_{12}^2 xy} = \sum_{nm} c_{nm} w^{1/2}(x) w^{1/2}(y) L_n(x) L_m(y), \quad (58)$$

where  $w(x)$  are the Gaussian quadrature weights and then determined the Schmidt vectors and coefficients by diagonalizing the matrix  $c_{nm}$ .<sup>30</sup> Figure 3(a) shows the resulting entropy for this limit as a function of the fluctuation strength  $\sigma_{12}^2$ . Interestingly, this shows



**FIG. 3.** (a) Entropy vs cross correlation magnitude  $\sigma_{12}$  in the case of slow modulation. (b) Schmidt modes for a maximally entangled state ( $\sigma_{12}^2 = 1.33$ ).

a maximum in the entanglement for  $\sigma_{12}^2 \approx 1.33$ . This can be understood in the following way. In the limit that  $\sigma_{12}^2$  is small, fluctuations are simply too weak to generate entanglement. On the other hand, large fluctuations also appear to prevent the formation of entangled states. The maximum then falls in the limit of being neither too soft nor too hard.<sup>31</sup> Figure 3(b) shows the Schmidt basis functions for the maximal entropy case where  $\sigma_{12}^2$ .

An interesting case arises when

$$\mathcal{K}^{(2)} = a\delta(t - t'). \quad (59)$$

Since one can write the delta-function as a resolution of the identity in terms of orthogonal polynomials  $O_n(x)$ ,<sup>30</sup>

$$\delta(t - t') = \lim_{N \rightarrow \infty} \sum_{k=0}^N O_k(t)O_k(t'), \quad (60)$$

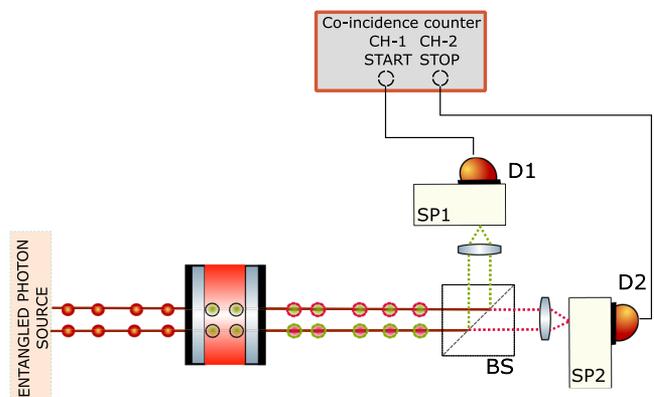
its normalized Schmidt coefficients are all equal to  $1/\sqrt{N}$ . This gives rise to the case of maximal entanglement entropy since

$$S = - \lim_{N \rightarrow \infty} \sum_{k=0}^N \frac{1}{N} \ln(1/N) \rightarrow \infty. \quad (61)$$

### III. DISCUSSION

To put into context the theoretical notions developed in this work, Fig. 4 sketches an envisioned experimental scheme that would quantify quantum entropy in analogy to that determined by Eq. (57). Time-frequency entangled photon pairs can be readily produced by spontaneous parametric downconversion of a continuous-wave laser source, and these are directed to an optical microcavity containing an absorber—either molecular materials or a semiconductor. Strong exciton-photon coupling leads to entangled polariton pairs. By this scheme, and depending on the photon-exciton detuning designed into the microcavity, the entangled photon pair will mix with the exciton to generate entangled polaritons that are either predominantly photonlike (negative detuning), excitonlike (positive detuning), or an equal mixture of photon/exciton (zero detuning). This specific character will guide the many-body response function  $\mathcal{S}^{(2)}(t_1, t_2, t'_1, t'_2)$ , which will determine the nature of the output photon state. The photon output is split by a beam splitter, and

each stage is directed to a spectrometer containing a single-photon avalanche diode, which permits measurement of coincidence count rates (rate of both detectors measuring a photon count within a short time window with respect to the detector response time). This count rate quantifies the biphoton scattering amplitudes. By this scheme, one can measure the coincidence rate as a function of the incidence angle and thus  $\vec{k}$  of the polariton dispersion. Spectrally resolved coincidence count rates can be used to characterize the joint spectral properties of the biphoton state, which is a measurement of its purity.<sup>32</sup> Performing such a measurement with and without the microcavity sample will permit estimation of changes in the entangled biphoton quantum state induced by matter-matter correlations. Slow (Gaussian) correlations between polariton modes facilitating the entanglement can be achieved by introducing an energy disorder for the quantum emitters. For example, one can use solvated chromophores or semiconductor quantum dots in polymer matrices.



**FIG. 4.** Proposed experimental implementation of an entanglement entropy measurement (spectrally resolved Hanbury Brown and Twiss experimental setup). BS = beam splitter; SP1, SP2 = spectrometers; and D1, D2 = single-photon detectors. Spectrally resolved coincidence count rates are measured to construct a photon spectral correlation coincidence map, from which the photon-photon scattering function  $\mathcal{S}^{(2)}(\omega_1, \omega_2, \omega'_1, \omega'_2)$  is reconstructed, thereby the entanglement entropy [in analogy to Eq. (57)] can be quantified.

We have presented here a model for the generation of entanglement entropy for a biphoton Fock state interacting with a material sample. We assume that the two-photon scattering matrix can be decomposed into a series of single photon/photon interactions mediated by coupling to a medium with coupling strength  $\xi$ . In the limit that the scattering produces a random phase shift, the entanglement collapses and the outgoing state is a single Fock state. However, in the case of Gaussian noise, the entanglement entropy increases with increasing coupling producing squeezed states. We also present a microscopic model for the photon-photon coupling for the case of two photons passing through an optical cavity. Here, we again show that in the limit of fast fluctuations and motional narrowing, the entanglement entropy vanishes, whereas in the case of slow modulation (homogeneous broadening), the entanglement entropy reaches a maximum value depending on the magnitude of the fluctuations. Our analysis shows that two-photon entanglement scattering provides a direct and sensitive probe of correlated fluctuations within the sample system.

## ACKNOWLEDGMENTS

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## APPENDIX A: SCHMIDT DECOMPOSITION OF EQ. (29)

Equation (29) is in the form of a Schmidt decomposition with coefficients

$$r_n = \xi^n e^{-n^2 \sigma^2 / 2} = e^{n \ln \xi - n^2 \sigma^2}. \quad (\text{A1})$$

From this, one can readily compute its von Neumann entropy

$$S = -\frac{1}{\Omega} \sum_n r_n^2 (\ln r_n^2 - \ln \Omega) \quad (\text{A2})$$

$$= \ln \Omega - 2m_1 \ln \xi + \sigma^2 m_2, \quad (\text{A3})$$

where  $\Omega$  provides the normalization

$$\sum_n r_n^2 = \Omega.$$

The sums can be evaluated as Gaussian integrals giving

$$\Omega = \frac{\sqrt{\pi} e^{-\frac{\ln^2(\xi)}{4\sigma^2}} \left( \operatorname{erf}\left(\frac{\ln(\xi)}{2\sqrt{\sigma^2}}\right) + 1 \right)}{2\sqrt{\sigma^2}}, \quad (\text{A4})$$

$$m_1 = \frac{e^{-\frac{\ln^2(\xi)}{4\sigma^2}}}{\sqrt{\pi}\sqrt{\sigma^2} \left( \operatorname{erf}\left(\frac{\ln(\xi)}{2\sqrt{\sigma^2}}\right) + 1 \right)} + \frac{\ln(\xi)}{2\sigma^2}, \quad (\text{A5})$$

$$m_2 = \frac{\sqrt{\sigma^2} e^{-\frac{\ln^2(\xi)}{4\sigma^2}} \left( \frac{\sqrt{\pi} \ln(\xi) e^{-\frac{\ln^2(\xi)}{4\sigma^2}} \left( \operatorname{erf}\left(\frac{\ln(\xi)}{2\sqrt{\sigma^2}}\right) + 1 \right)}{(\sigma^2)^{3/2}} + \frac{2}{\sigma^2} \right)}{2\sqrt{\pi} \left( \operatorname{erf}\left(\frac{\ln(\xi)}{2\sqrt{\sigma^2}}\right) + 1 \right)} \quad (\text{A6})$$

assuming that  $\ln \xi < 0$  and  $\operatorname{Re}[\sigma^2] > 0$ .

## APPENDIX B: PHOTON MODE SCATTERING VIA CAVITY POLARITON FLUCTUATIONS

In this appendix, we derive a stochastic Hamiltonian (35) based on a simple model describing light scattering by the fluctuations of delocalized polariton modes confined in an optical cavity. For this situation, the photon Hamiltonian can be represented as a sum of two components

$$\hat{H}_{\text{ph}} = \hat{H}_o + \hat{H}_s. \quad (\text{B1})$$

Assuming that the biphoton wavepacket is spatially confined within a cross section of area  $A$  and propagates in the  $z$ -direction, the interaction free photon Hamiltonian in the continuous mode representation reads<sup>24</sup>

$$\hat{H}_o = \sum_{j=1,2} \int d\omega_j \omega_j \hat{b}_{\omega_j}^\dagger \hat{b}_{\omega_j}, \quad (\text{B2})$$

with index  $j$  distinguishing the modes.

The photon scattering is described by the interaction Hamiltonian<sup>29</sup>

$$\hat{H}_s = - \iint_A dx dy \int_{-L/2}^{L/2} dz \sum_{j=1,2} \hat{\alpha}_{ij}(\mathbf{r}) \hat{E}_j^2(z), \quad (\text{B3})$$

where the integration  $dS$  over the photon wave packet cross section ( $x, y$ -plane) is partitioned from the spatial integral in the propagation direction  $z$ . The operator  $\hat{\alpha}_{ij}(\mathbf{r})$  gives the sample polarizability. The cavity cross section is assumed to be larger than  $A$  and the length is denoted by  $L$ . The electric field operator for the photon modes of interest represented in terms of the mode creation and annihilation operators reads<sup>24</sup>

$$\hat{E}_j(z) = i \int d\omega_j \sqrt{\frac{\hbar \omega_j}{4\pi \epsilon_o c A}} \left( \hat{b}_{\omega_j} e^{i\omega_j z/c} - \hat{b}_{\omega_j}^\dagger e^{-i\omega_j z/c} \right), \quad (\text{B4})$$

where  $c$  is speed of light and  $\epsilon_o$  is the vacuum permittivity.

Evaluation of the integrals in the scattering Hamiltonian (B3) requires a model for the sample polarizability operator  $\hat{\alpha}(\mathbf{r})$ . Let us consider delocalized cavity polariton modes which we described by operator

$$\hat{\zeta}_{\mathbf{k}_l} = \bar{\zeta}_{\mathbf{k}_l} + \delta \hat{\zeta}_{\mathbf{k}_l}(t), \quad (\text{B5})$$

where  $\mathbf{k}_l$  denotes sth polariton mode wave vector.  $\bar{\zeta}_{\mathbf{k}_l}$  is a cavity polariton steady state prepared by a resonant external pumping and  $\delta \hat{\zeta}_{\mathbf{k}_l}(t)$  is the time-dependent mode fluctuation operator. Accordingly, the polarizability can be expanded up to the first order in the fluctuations

$$\hat{\alpha}_{jj} = \alpha_{jj}(\bar{\zeta}_{\mathbf{k}_l}) + \sum_l \sum_{\mathbf{k}_l} \frac{\partial \alpha_{jj}(\bar{\zeta}_{\mathbf{k}_l})}{\partial \bar{\zeta}_{\mathbf{k}_l}} \delta \hat{\zeta}_{\mathbf{k}_l}(t). \quad (\text{B6})$$

For further analysis, the fluctuation operator is expanded in terms of polariton spatial Fourier components

$$\delta\hat{\zeta}_{\mathbf{k}_l}(t) = \delta\hat{\zeta}_{\mathbf{k}_l}^+(t)e^{i\mathbf{k}_l r} + \delta\hat{\zeta}_{\mathbf{k}_l}^-(t)e^{-i\mathbf{k}_l r}. \quad (\text{B7})$$

Making substitution of the second term in Eq. (B6) along with Eq. (B7) into the scattering Hamiltonian (B3) where the electric field is introduced by Eq. (B4), performing integration over the cavity volume, and further neglecting the terms describing simultaneous two-photon creation and annihilation processes, we obtain

$$\hat{H}_s = -\hbar \sum_l \sum_{\mathbf{k}_l} \sum_{j=1,2} \int d\omega_j \kappa_{\mathbf{k}_l}(\omega_j) \left( \hat{b}_{\omega_j+c\mathbf{k}_l}^\dagger \hat{b}_{\omega_j} \delta\hat{\zeta}_{\mathbf{k}_l}^+ + \hat{b}_{\omega_j}^\dagger \hat{b}_{\omega_j+c\mathbf{k}_l} \delta\hat{\zeta}_{\mathbf{k}_l}^- \right), \quad (\text{B8})$$

with the coupling parameter

$$\kappa_{\mathbf{k}_l}(\omega_j) = \frac{1}{\epsilon_0 L} \left. \frac{\partial \alpha_{jj}(\tilde{\zeta}_{\mathbf{k}_l})}{\partial \tilde{\zeta}_{\mathbf{k}_l}} \right|_{\mathbf{k}_l^\perp=0} [(\omega_j - c\mathbf{k}_l)\omega_j]^{1/2}. \quad (\text{B9})$$

Since the photons propagate in the  $z$ -direction, the total momentum conservation requires that the scattered photon momentum changes for the amount of  $k_j$  which is the  $z$ -component of the total momentum  $\mathbf{k}_j$ . Accordingly, the transverse ( $xy$ -plane) component of the momentum  $\mathbf{k}_j^\perp$  does not change resulting in the photon coupling to the  $\Gamma$ -point of transverse polariton band as indicated above by setting  $\mathbf{k}_j^\perp = 0$ .

Taking into account that polariton modes have continuous dispersion relations  $k_l = k(\omega_l)$ , we replace sum over  $\mathbf{k}_l$  in Eq. (B8) by the integral over  $d\omega_l$ . This results in

$$\hat{H}_s = -\hbar \sum_{j=1,2} \sum_l \int \int d\omega_j d\omega_l \kappa(\omega_j \omega_l) \left( \hat{b}_{\omega_j+\omega_l}^\dagger \hat{b}_{\omega_j} \delta\hat{\zeta}_{\omega_l}^+ + \hat{b}_{\omega_j}^\dagger \hat{b}_{\omega_j+\omega_l} \delta\hat{\zeta}_{\omega_l}^- \right), \quad (\text{B10})$$

with the coupling parameter

$$\kappa(\omega_j \omega_l) = \frac{1}{2\pi\epsilon_0} \left. \frac{\partial \alpha_{jj}(\tilde{\zeta}_{\mathbf{k}(\omega_l)})}{\partial \tilde{\zeta}_{\mathbf{k}(\omega_l)}} \right|_{\mathbf{k}^\perp(\omega_l)=0} \frac{\partial k(\omega_l)}{\partial \omega_l} [(\omega_j - \omega_l)\omega_j]^{1/2}. \quad (\text{B11})$$

The scattering Hamiltonian (B10) can be further simplified, provided the interaction occurs near the bottom of polariton modes, i.e.,  $\omega_l \sim 0$ . In this case, one can set

$$\left. \frac{\partial \alpha_{jj}(\tilde{\zeta}_{\mathbf{k}(\omega_l)})}{\partial \tilde{\zeta}_{\mathbf{k}(\omega_l)}} \right|_{\mathbf{k}^\perp(\omega_l)=0} = \tilde{\alpha}_{jj} \delta(\omega_l) \quad (\text{B12})$$

with  $\tilde{\alpha}_{jj}$  being a coupling constant. Substitution of Eq. (B11) with Eq. (B12) into Eq. (B10) recasts the latter to the form of stochastic Hamiltonian (35) with the frequency fluctuation operator defined as

$$\delta\hat{\omega}_j = \frac{\tilde{\alpha}_{jj}\omega_j}{2\pi\epsilon_0} \sum_l \frac{\partial k(\omega_l)}{\partial \omega_l} \delta\hat{\zeta}_l, \quad (\text{B13})$$

where a shorthand notation  $\delta\hat{\zeta}_l = \delta\hat{\zeta}_{\omega_l=0}$  is used.

## APPENDIX C: EVALUATION OF SECOND CUMULANTS FOR GAUSSIAN STOCHASTIC PROCESS

In this appendix, we derive an explicit form of the single-mode and cross-mode cumulant functions using the correlation functions for stochastic Gaussian processes given in Eqs. (48) and (49), respectively.

Evaluation of time ordered integral in Eq. (46) with the correlation function given by Eq. (48) results in a well known form of the second cumulant<sup>28</sup>

$$g_i(t-t') = (\sigma_j \bar{\tau}_j)^2 \left\{ (t-t')/\bar{\tau}_j + e^{-(t-t')/\bar{\tau}_j} + 1 \right\}. \quad (\text{C1})$$

In the case of fast modulation  $\sigma \bar{\tau}_j \ll 1$ , one gets

$$g_j(t-t') = \gamma_j(t-t'), \quad (\text{C2})$$

with  $\gamma_j = \sigma^2 \bar{\tau}_j$ . In the case of fast modulation  $\sigma \bar{\tau}_j \gg 1$ , one gets

$$g_i(t-t') = \sigma_j^2 (t-t')^2. \quad (\text{C3})$$

Evaluation of the integrals in Eq. (47) with the cross correlation function in Eq. (49) is not so straightforward and one needs to take into account various ordering of  $t'_1, t_1, t'_2$ , and  $t_2$  as sketched in Fig. 5 with three more corresponding to swapping indices 1 and 2 (but not the primes)

$$g_{12}^+(t_1 t'_1 t_2 t'_2) = (\sigma_{12} \bar{\tau}_{12})^2 \left\{ e^{-(t_1-t_2)/\bar{\tau}_{12}} - e^{-(t_1-t'_2)/\bar{\tau}_{12}} \times e^{-(t'_1-t_2)/\bar{\tau}_{12}} - e^{-(t'_1-t'_2)/\bar{\tau}_{12}} \right\}, \quad (\text{C4})$$

$$g_{12}^+(t_1 t_2 t'_1 t'_2) = (\sigma_{12} \bar{\tau}_{12})^2 \left\{ 2(t_2 - t'_1)/\bar{\tau}_{12} e^{-(t_1-t_2)/\bar{\tau}_{12}} + e^{-(t_1-t'_2)/\bar{\tau}_{12}} e^{-(t'_1-t_2)/\bar{\tau}_{12}} + e^{-(t_2-t'_1)/\bar{\tau}_{12}} \right\}, \quad (\text{C5})$$

$$g_{12}^+(t_1 t_2 t'_2 t'_1) = (\sigma_{12} \bar{\tau}_{12})^2 \left\{ 2(t_2 - t'_2)/\bar{\tau}_{12} e^{-(t_2-t'_1)/\bar{\tau}_{12}} + e^{-(t_2-t'_1)/\bar{\tau}_{12}} e^{-(t_1-t_2)/\bar{\tau}_{12}} + e^{-(t_1-t'_2)/\bar{\tau}_{12}} \right\} \quad (\text{C6})$$

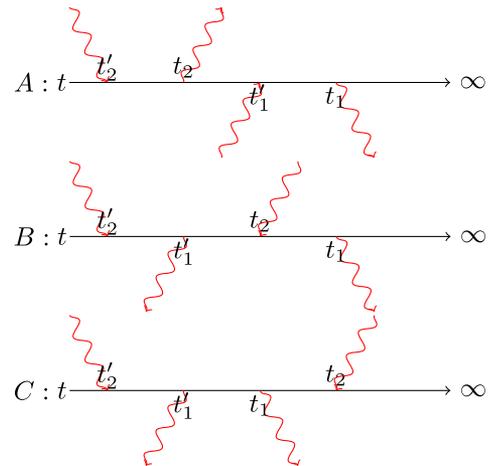


FIG. 5. (A)–(C): Time ordered interactions corresponding to Eqs. (C4)–(C6).

and three more

$$g_{12}^+(t_2 t_1 t'_1 t'_2) = (\sigma_{12} \bar{\tau}_{12})^2 \left\{ 2(t_1 - t'_1) / \bar{\tau}_{12} e^{-(t'_1 - t'_2) / \bar{\tau}_{12}} + e^{-(t_1 - t'_2) / \bar{\tau}_{12}} e^{-(t_2 - t_1) / \bar{\tau}_{12}} + e^{-(t_2 - t'_1) / \bar{\tau}_{12}} \right\}, \quad (C7)$$

$$g_{12}^+(t_2 t_1 t'_2 t'_1) = (\sigma_{12} \bar{\tau}_{12})^2 \left\{ 2(t_1 - t'_2) / \bar{\tau}_{12} e^{-(t_2 - t_1) / \bar{\tau}_{12}} + e^{-(t_2 - t'_1) / \bar{\tau}_{12}} + e^{-(t_1 - t'_2) / \bar{\tau}_{12}} \right\}, \quad (C8)$$

$$g_{12}^+(t_2 t'_2 t_1 t'_1) = (\sigma_{12} \bar{\tau}_{12})^2 \left\{ e^{-(t_2 - t_1) / \bar{\tau}_{12}} - e^{-(t_2 - t'_1) / \bar{\tau}_{12}} \right. \\ \left. \times e^{-(t'_2 - t_1) / \bar{\tau}_{12}} - e^{-(t'_2 - t'_1) / \bar{\tau}_{12}} \right\}, \quad (C9)$$

with time indices 1 and 2 swapped. Note, our notation is such that in  $g_{12}^+(t_a t_b t_c t_d)$ ,  $t_a > t_b > t_c > t_d$ .

In the case of *fast modulation*  $\sigma_{12} \bar{\tau}_{12} \ll 1$ ,  $g_{12}^+(t_1 t'_1 t_2 t'_2) = 0$ ,  $g_{12}^+(t_2 t'_2 t_1 t'_1) = 0$ , and the rest of time-ordered cumulants simplify to the form

$$g_{12}^+(t_1 t_2 t'_1 t'_2) = \gamma_{12}(t_2 - t'_1), \quad (C10)$$

$$g_{12}^+(t_1 t_2 t'_2 t'_1) = \gamma_{12}(t_2 - t'_2), \quad (C11)$$

$$g_{12}^+(t_2 t_1 t'_2 t'_1) = \gamma_{12}(t_1 - t'_2), \quad (C12)$$

$$g_{12}^+(t_2 t_1 t'_1 t'_2) = \gamma_{12}(t_1 - t'_1), \quad (C13)$$

with  $\gamma_{12} = \sigma_{12}^2 \bar{\tau}_{12}$ .

In the case of *slow modulation*  $\sigma_{12} \bar{\tau}_{12} \gg 1$ , Eqs. (C4)–(C9) simplify to the following expression:

$$g_{12}(t - t') = \sigma_{12}^2 (t_1 - t'_1)(t_2 - t'_2), \quad (C14)$$

which holds for all initial time permutations used in Eqs. (C4)–(C9).

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