Correlation of photon pairs from the double Raman amplifier: Generalized analytical quantum Langevin theory

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We present a largely analytical theory for two-photon correlations $G^{(2)}$ between Stokes (s) and anti-Stokes (a) photon pairs from an extended medium (amplifier) composed of double-Λ atoms in counterpropagating geometry. We generalize the parametric coupled equations with quantum Langevin noise given in a beautiful experimental paper of Balic *et al.* [Phys. Rev. Lett. **94**, 183601 (2005)] beyond adiabatic approximation and valid for arbitrary strength and detuning of laser fields. We derive an analytical formula for cross correlation $G_{as}^{(2)}=\langle \hat{E}_s^\dagger(L)\hat{E}_a^\dagger(0,\tau)\hat{E}_a(0,\tau)\hat{E}_s(L)\rangle$ and use it to obtain results that are in good quantitative agreement with the experimental data. Results for $G_{as}^{(2)}$ obtained using our coupled equations are in good quantitative agreement with the results using the equations of Balic *et al.*, while perfect agreement is obtained for sufficiently large detuning. We also compute the reverse correlation $G_{sa}^{(2)}$ which turns out to be negligibly small and remains classical while the cross correlation violates the Cauchy-Schwartz inequality by a factor of more than a hundred.

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I. INTRODUCTION

One of the amazing properties of the quantum world is quantum correlation. The quantum mechanical concept of photon-photon correlation introduced by Glauber has provided us with insights into the distinct quantum statistical nature of photons from various light sources such as lasers, the Sun, and resonance fluorescence. Spontaneous emission, being a quantum mechanical process, has an important role in establishing quantum correlation. Recently, various versions of double-Λ schemes have been explored [1] in regards to entanglement, but not so much in the interest of quantum correlation. Entanglement, the heart of quantum informatics, is related to quantum correlation, a concept which extends beyond the pure state.

Furthermore, quantum correlation in the sense of Glauber's two-photon correlation $G^{(2)}$ in an extended medium with propagation deserves proper theoretical studies, especially for the double-Λ scheme. The scheme has remarkable features and has been widely studied in the context of quantum erasers [2], quantum information [3–5], nonlinear optics [6,7], and subwavelength resolution microscopy [8]. A Stokes photon is generated via a spontaneous Raman process. It is possible to generate another photon, the anti-Stokes photon, which is strongly correlated to the Stokes photon by applying a strong resonant control field Ω_c [Fig. 1(a)]. The laser field creates a dressed state $\frac{1}{\sqrt{2}}(|a,n_c-1\rangle+|b,n_c\rangle)$ (with n_c the photon number of the control field): a coherent superposition of state $|b,n_c\rangle$ following the emission of a Stokes photon and state $|a,n_c-1\rangle$ from which an anti-Stokes photon would be emitted. We refer to the correlated photon pairs produced by this "spontaneous Raman-EIT (electromagnetically induced transparency)" scheme as the *Raman emission doublet* (RED). The scheme exhibits nonclassical properties such as squeezing [6], violation of the Cauchy-Schwartz inequality [3,9], and antibunching with Rabi oscillations in $G^{(2)}$ for the single-atom case [10]. The RED scheme also enables efficient mapping of the quantum information of the input Stokes photon into the atomic ensembles and reading off the information as an anti-Stokes photon after a controllable time delay up to 2 μs [5], much longer than those produced in cascade scheme and in parametric down-conversion [11].

In an extended medium where a large number of correlated photon pairs can be generated, application of the scheme in quantum lithography [12] becomes more feasible. The correlation time can be increased via a slow light effect through the control field. Recently, nonclassical macroscopic photon correlation of the RED scheme has been demonstrated for many (cold ^{87}Rb) atoms in backward propagating geometry [13] in a beautiful experiment by Balic *et al.* [9]. They obtained coupled parametric oscillator equations based on the adiabatic approximation which assumes that the entire population is in the ground level. Their experimental data were fitted remarkably well with the numerical solutions of the coupled equations.

Motivated by their work, here we extend their coupled equations beyond the RED scheme. Without the adiabatic approximation, we derive the coupled parametric equations that are valid for any detuning and strength of the pump and control laser fields and arbitrary populations. We have obtained analytical expressions for the two-photon correlation in an extended medium with noise operators (more specifically the cross correlation $G_{as}^{(2)}$, reverse correlation $G_{sa}^{(2)}$, and self-correlations $G_{ff}^{(2)}$, with $f=s,a$) which seems to be a formidable task so far. The results for $G_{as}^{(2)}$ [Eq. (28)] are in

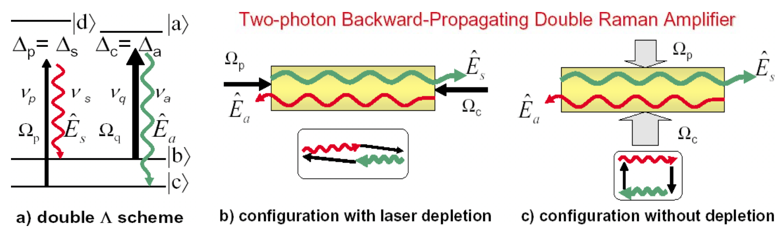

FIG. 1. (Color online) (a) The double Raman scheme produces correlated pairs of Stokes and anti-Stokes photons. The present four-level scheme (instead of three levels) describes the experimental situation in [9]. The coupling of the pump and Stokes fields to level $|a\rangle$ is negligible since the fields are far detuned from it. The Raman emission doublet (RED) scheme corresponds to $\Delta_{p,s} \gg \gamma_{ac}$, Ω_p, $\Delta_{c,a}=0$, and $\Omega_c > \Omega_p$ where the Stokes photon is generated by the spontaneous Raman process and the anti-Stokes is by the resonant Raman process. Counterpropagating Stokes and anti-Stokes photons amplified into correlated macroscopic quantum fields $\hat{E}_s$ and $\hat{E}_a$ are generated by lasers propagating (b) along the extended medium (laser depletion may be significant) and (c) perpendicular to the extended medium (negligible laser depletion). Also shown are phase-matched diagrams of the four fields.

good agreement with the experiment [9]. On the other hand, we find that the $G_{sa}^{(2)}$ is negligibly small for the RED scheme, as expected [15].

Even though we focus on the small signal regime and disregard laser field depletion, our theory yields a lot of new results for laser parameters in different limiting cases. Further results and analysis will be reported in a series of forthcoming papers. They include (a) forward (copropagating) geometry, (b) the role of noise operators and the connection with group delay in signal propagation [16], and (c) dependence of nonclassicalness on laser parameters.

In Sec. II, we present the coupled oscillator equations for counterpropagating (backward geometry) laser fields with arbitrary strength and detuning. The solutions of the coupled equations in the frequency domain for the boundary parts and the noise parts are given in Sec. III. We show that they reduce to the known solutions in Ref. [6] in the case of the RED scheme. In Sec. IV, we derive analytical expressions for $G_{as}^{(2)}$, $G_{sa}^{(2)}$, and $G_{ff}^{(2)}$. Normalized correlations such as Cauchy-Schwartz correlation are defined in Sec. V to quantify the degree of nonclassicalness. We also compute the quantitative detection rate of the experiment [9] from $G_{as}^{(2)}$. Finally, in Sec. VI we compare the computed detection rates (based on our coupled equations and analytical correlation) and the hybrid results (the coupled equations of Balic *et al.* and our analytical correlation) with the experimental data [9].

II. GENERALIZED COUPLED EQUATIONS FOR EXTENDED MEDIUM

We proceed to the main focus of this paper: to study the two-photon correlation between the Stokes and anti-Stokes fields of an extended medium or amplifier composed of a macroscopic number of atoms with the double Raman scheme [Fig. 1(a)]. Despite the appreciable propagation effect in the extended sample, the off-resonant and weak pump field in the RED scheme lead to a negligible depletion of the laser fields even for the scheme in Fig. 1(b). It is possible to minimize the depletion of laser fields via the configuration where the lasers are orthogonal to the Stokes and anti-Stokes fields [Fig. 1(c)]. This enables us to neglect Maxwell's equations for the laser fields which describe the depletion. The resulting Maxwell's equations for the macroscopic [17] Stokes $\hat{E}_a=\hat{A}/g_a$ and anti-Stokes $\hat{E}_s=\hat{S}/g_s$ fields ($g_s=\wp_{db}/\hbar$ and $g_a=\wp_{ac}/\hbar$ as the coupling strengths) together with linearized atomic equations (Appendix A) lead to [21] the coupled equations

$$\left(\frac{\partial}{\partial z}+G_s\right)\hat{S}+\left(K_s-\beta_s\frac{\partial}{\partial z}\right)\hat{A}^\dagger=\hat{F}_s, \tag{1}$$

$$\left(-\frac{\partial}{\partial z}+G_a\right)\hat{A}^\dagger+\left(K_a+\beta_a\frac{\partial}{\partial z}\right)\hat{S}=\hat{F}_a^\dagger, \tag{2}$$

with the effective noise operators

$$\hat{F}_s=X_{ad}\hat{G}_{ad}+X_{bc}\hat{G}_{bc}+X_{bd}\hat{G}_{bd}, \tag{3}$$

$$\hat{F}_a^\dagger=Y_{ad}\hat{G}_{ad}+Y_{bc}\hat{G}_{bc}+Y_{ac}\hat{G}_{ac}, \tag{4}$$

where the coefficients G_f, K_f, β_f, X_x, and Y_x ($x=ac,ad,bc,bd$) are given by Eqs. (B1)–(B4) and (B7)–(B10) in Appendix B, respectively, and $\hat{G}_x=\int_{-\infty}^{\infty}e^{i\theta_x}\hat{F}_x(z,t)e^{i\nu t}dt$ is the Fourier transform of the noise operators in Eqs. (A6)–(A9) including the rapidly varying phases $\theta_x(z,t)=k_x z-\nu_x t$. Equations (1) and (2) describe the dynamical evolutions of the macroscopic quantum fields for the Stokes and anti-Stokes photons in the spectral domain.

By multiplying Eq. (1) by β_a and then subtracting it from Eq. (2) and similarly multiplying Eq. (2) by β_s and subtracting it from Eq. (1) we obtain the familiar *parametric oscillator* coupled equations [9,22]

$$\left(\frac{\partial}{\partial z}+\mathcal{G}_s\right)\hat{S}+\mathcal{K}_s\hat{A}^\dagger=\bar{F}_s, \tag{5}$$

$$\left(-\frac{\partial}{\partial z}+\mathcal{G}_a\right)\hat{A}^\dagger+\mathcal{K}_a\hat{S}=\bar{F}_a^\dagger, \tag{6}$$

since the coefficients in Eqs. (1) and (2) are related to that of Eqs. (5) and (6) by

$$\mathcal{G}_s = \frac{(G_s - \beta_s K_a)}{I_{sa}}, \quad \mathcal{G}_a = \frac{(G_a - \beta_a K_s)}{I_{sa}}, \tag{7}$$

$$\mathcal{K}_s = \frac{(K_s - \beta_s G_a)}{I_{sa}}, \quad \mathcal{K}_a = \frac{(K_a - \beta_a G_s)}{I_{sa}}, \tag{8}$$

$$\bar{F}_s = \frac{\hat{F}_s - \beta_s \hat{F}_a^\dagger}{I_{sa}}, \quad \bar{F}_a^+ = \frac{\hat{F}_a^\dagger - \beta_a \hat{F}_s}{I_{sa}}, \tag{9}$$

with $I_{sa}=1-\beta_s\beta_a$. The two sets of coefficients are approximately equal when $\beta_s \simeq \beta_a \simeq 0$. Here, the $\mathcal{G}_s$ is the spontaneous Raman gain coefficient, $\mathcal{G}_a$ gives the EIT dispersion and absorption profiles modified in the presence of the pump laser, and $\mathcal{K}_s$ and $\mathcal{K}_a$ are the cross couplings. The coefficients $\mathcal{G}_f$ and $\mathcal{K}_f$ generalize those obtained by Refs. [9,22] beyond the adiabatic approximation.

The effective Fourier transforms of the noise operators $\bar{F}_s$ and $\bar{F}_a^\dagger$ serve as the driving "forces" or seeds to both fields and their physical origin is the quantum vacuum fluctuations.

Note that Eqs. (5) and (6) are equivalent to a driven oscillator equation with effective gain or damping $(\mathcal{G}_s+\mathcal{G}_a)$ and oscillation angular frequency $\sqrt{\mathcal{G}_a\mathcal{G}_s-\mathcal{K}_s\mathcal{K}_a}$. These coupled equations are also obtained for the case of the resonance fluorescence of two-level atoms in an extended medium, which will be a subject of future publications.

III. SOLUTIONS FOR COUNTERPROPAGATING (BACKWARD) GEOMETRY

The solutions of the generalized coupled equations for the Stokes field at $z=L$ and the anti-Stokes field at $z=0$ are composed of the boundary operators $\hat{B}_f$ (specifically the Stokes operator at $z=0$ and the anti-Stokes operator at $z=L$) and noise operators $\hat{N}_f$ [specifically $\hat{F}_f(z,\nu)$ at all points in the medium]—i.e.,

$$\hat{E}_s(L,\nu) = \underbrace{\bar{\psi}_s^s(L,\nu)\hat{E}_s(0,\nu) + \bar{\psi}_a^s(L,\nu)\frac{g_a^*}{g_s}\hat{E}_a^\dagger(L,\nu)}_{\hat{B}_s(L,\nu)\text{ boundary Stokes}} + \underbrace{\frac{1}{g_s}\int_0^L [\bar{U}_s^s(\xi,\nu)\hat{F}_s(z,\nu) + \bar{U}_a^s(\xi,\nu)\hat{F}_a^\dagger(z,\nu)]dz}_{\hat{N}_s(L,\nu)\text{ noise Stokes}}, \tag{10}$$

$$\hat{E}_a^\dagger(0,\nu) = \underbrace{\bar{\psi}_a^a(L,\nu)\hat{E}_a^\dagger(L,\nu) + \bar{\psi}_s^a(L,\nu)\frac{g_s}{g_a^*}\hat{E}_s(0,\nu)}_{\hat{B}_a(L,\nu)\text{ boundary anti-Stokes}} + \underbrace{\frac{1}{g_a^*}\int_0^L [\bar{U}_a^a(\xi,\nu)\hat{F}_a^\dagger(z,\nu) + \bar{U}_s^a(\xi,\nu)\hat{F}_s(z,\nu)]dz}_{\hat{N}_a(L,\nu)\text{ noise anti-Stokes}} \tag{11}$$

where coefficients for the boundary operators are

$$\bar{\psi}_s^s(L,\nu) = \Xi_{\bar{q}}(L,\nu) + \Xi(L,\nu)\{\mathcal{G}_a + Z(L,\nu)\mathcal{K}_a\}, \tag{12}$$

$$\bar{\psi}_a^s(L,\nu) = Z(L,\nu), \tag{13}$$

$$\bar{\psi}_a^a(L,\nu) = \frac{1}{\Xi_{\bar{q}}(L,\nu) - \Xi(L,\nu)\mathcal{G}_s}, \tag{14}$$

$$\bar{\psi}_s^a(L,\nu) = \frac{\mathcal{K}_a}{\mathcal{K}_s}Z(L,\nu), \tag{15}$$

with

$$I_{sa} = 1 - \beta_s\beta_a, \tag{16}$$

$$Z(L,\nu) = \frac{\Xi(L)\mathcal{K}_s}{\Xi_{\bar{q}}(L) - \Xi(L)\mathcal{G}_s}, \tag{17}$$

the oscillatory functions

$$\Xi_{\bar{q}}(x,\nu) = \frac{\bar{q}_+ e^{-\bar{q}_+ x} - \bar{q}_- e^{-\bar{q}_- x}}{\bar{q}_+ - \bar{q}_-}, \tag{18}$$

$$\Xi(x,\nu) = \frac{e^{-\bar{q}_+ x} - e^{-\bar{q}_- x}}{\bar{q}_+ - \bar{q}_-}, \tag{19}$$

and effective wave vectors

$$\bar{q}_\pm = -\bar{\alpha} \pm \bar{\beta}, \tag{20}$$

$$\bar{\alpha} = \frac{1}{2}(\mathcal{G}_a - \mathcal{G}_s), \tag{21}$$

$$\bar{\beta} = \sqrt{\bar{\alpha}^2 - (\mathcal{K}_s\mathcal{K}_a - \mathcal{G}_s\mathcal{G}_a)}. \tag{22}$$

The kernels for the noise operators are

$$\bar{U}_s^s(\xi,\nu) = \frac{1}{I_{sa}}[\Xi_{\bar{q}}(\xi)(1 - Z\beta_a) + \Xi(\xi)(G_a + ZK_a)], \tag{23}$$

$$\bar{U}_a^s(\xi,\nu) = \frac{1}{I_{sa}}[\Xi_{\bar{q}}(\xi)(Z - \beta_s) - \Xi(\xi)(K_s + ZG_s)], \tag{24}$$

$$\bar{U}_a^a(\xi,\nu) = \frac{\Xi_{\bar{q}}(\xi) - \Xi(\xi)G_s}{D(L)}, \tag{25}$$

$$\bar{U}_s^a(\xi,\nu) = -\frac{\beta_a\Xi_{\bar{q}}(\xi) - K_a\Xi(\xi)}{D(L)}, \tag{26}$$

where $D(L)=I_{sa}\{\Xi_{\bar{q}}(L)-\Xi(L)\mathcal{G}_s\}$. Equations (10) and (11) are general solutions for backward geometry with arbitrary detuning and laser fields.

The limit $\beta_s \simeq \beta_a \ll 1$ corresponds to the RED scheme where we find $\bar{U}_s^s(\xi,\nu) \to \bar{\psi}_s^s(\xi,\nu)$ and $\bar{U}_s^a(\xi,\nu) \to \bar{\psi}_s^a(\xi,\nu)$ but $\bar{U}_a^a(\xi,\nu) \nrightarrow \bar{\psi}_a^a(\xi,\nu)$ and $\bar{U}_a^s(\xi,\nu) \nrightarrow \bar{\psi}_a^s(\xi,\nu)$ in contrast with the case of forward geometry. However, in the limit of short samples, $\bar{q}_+ x \ll 1$, we have $\Xi_{\bar{q}} \simeq 1$, $\Xi \simeq 0$ and hence $\bar{U}_a^a(\xi,\nu) \to \bar{\psi}_a^a(\xi,\nu)$ and $\bar{U}_a^s(\xi,\nu) \to \bar{\psi}_a^s(\xi,\nu)$; i.e., the coefficients of the noise part are identical to that of the boundary part. This explains the correspondence of the results with boundary operators and that with noise operators.

We have verified that by neglecting β_f our Eqs. (10) and (11) reproduce the solutions of Ref. [6] (with the additional boundary operators) for the RED scheme.

IV. ANALYTICAL TWO-PHOTON CORRELATION

We now use the solutions for the field operators and the Glauber's two-photon correlation $G^{(2)}$ to compute the cross correlations between the Stokes and anti-Stokes fields, $G_{as}^{(2)}$ and $G_{sa}^{(2)}$, and the self-correlations $G_{ss}^{(2)}$ and $G_{aa}^{(2)}$. One way to derive the two-photon correlation is to note that it can be expressed as decorrelated [24] paired products

$$G_{as}^{(2)}(L,\tau) = |\langle \hat{E}_a(t+\tau)\hat{E}_s(t)\rangle|^2 + \langle \hat{E}_s^\dagger(t)\hat{E}_s(t)\rangle\langle \hat{E}_a^\dagger(t+\tau)\hat{E}_a(t+\tau)\rangle, \quad (27)$$

where the terms $\langle \hat{E}_s^\dagger(t)\hat{E}_a(t+\tau)\rangle$ and $\langle \hat{E}_a^\dagger(t+\tau)\hat{E}_s(t)\rangle$ vanish since $\langle \hat{N}_s^\dagger(t)\hat{N}_a(t+\tau)\rangle \propto \langle \hat{F}_x^\dagger(\nu)\hat{F}_{x'}^\dagger(\nu')\rangle = 0$ and $\langle \hat{N}_a^\dagger(t+\tau)\hat{N}_s(t)\rangle \propto \langle \hat{F}_x(\nu)\hat{F}_{x'}(\nu')\rangle = 0$.

Thus, the correlation for backward geometry can be computed as

$$G_{as}^{(2)}(\tau) = |\langle \hat{B}_a(L,t+\tau)\hat{B}_s(L,t)\rangle + \langle \hat{N}_a(L,t+\tau)\hat{N}_s(L,t)\rangle|^2 + \{\mathcal{I}_s^b(L) + \mathcal{I}_s^n(L)\}\{\mathcal{I}_a^b(L) + \mathcal{I}_a^n(L)\}, \quad (28)$$

where $\langle \hat{B}_a(L,t+\tau)\hat{B}_s(L,t)\rangle$, $\langle \hat{N}_a(L,\tau)\hat{N}_s(L)\rangle$, $\mathcal{I}_f^b(L,\tau)$, and $\mathcal{I}_f^n(L,\tau)$ are given below and are evaluated in a similar fashion in Appendix C for Eq. (C4). The superscripts "b" and "n" indicate that the terms are evaluated with boundary operators and noise operators, respectively.

For an inseparable two-photon *pure* state $|\Psi\rangle$, the correlation can be described by $G_{as}^{(2)}(L,\tau) = |\langle 0|\hat{E}_a(L,t+\tau)\hat{E}_s(L,t)|\Psi\rangle|^2$. Thus, the first term in Eq. (28) describes the correlation of the two-photon state while the second term describes the stimulated quantum fields corresponding to the uncorrelated states of two photons. For long time delay, the two photons become uncorrelated and $G_{as}^{(2)}$ would take a constant value given by the second term of Eq. (28) which is simply a direct product of the intensities of the Stokes and anti-Stokes photons.

The corresponding self-correlations are

$$G_{ss}^{(2)}(\tau) = \langle \hat{E}_s^\dagger(L,t)\hat{E}_s^\dagger(L,t+\tau)\hat{E}_s(L,t+\tau)\hat{E}_s(L,t)\rangle = |\mathcal{I}_s^b(L,\tau) + \mathcal{I}_s^n(L,\tau)|^2 + \{\mathcal{I}_s^b(L) + \mathcal{I}_s^n(L)\}^2, \quad (29)$$

$$G_{aa}^{(2)}(\tau) = \langle \hat{E}_a^\dagger(0,t)\hat{E}_a^\dagger(0,t+\tau)\hat{E}_a(0,t+\tau)\hat{E}_a(0,t)\rangle = |\mathcal{I}_a^b(L,\tau) + \mathcal{I}_a^n(L,\tau)|^2 + \{\mathcal{I}_a^b(L) + \mathcal{I}_a^n(L)\}^2. \quad (30)$$

Similarly, the reverse correlation for backward geometry is

$$G_{sa}^{(2)}(\tau) = \langle \hat{E}_a^\dagger(0,t)\hat{E}_s^\dagger(L,t+\tau)\hat{E}_s(L,t+\tau)\hat{E}_a(0,t)\rangle = |\langle \hat{B}_s(L,t+\tau)\hat{B}_a(L,t)\rangle + \langle \hat{N}_s(L,t+\tau)\hat{N}_a(L,t)\rangle|^2 + \{\mathcal{I}_s^b(L) + \mathcal{I}_s^n(L)\}\{\mathcal{I}_a^b(L) + \mathcal{I}_a^n(L)\}. \quad (31)$$

A. Noise products

The cross-correlation amplitude in Eq. (28) due to noise is derived in Appendix C as

$$\langle \hat{N}_a(L,\tau)\hat{N}_s(L)\rangle = e^{i\Delta kL}\frac{2\pi}{AN}\int_{-\infty}^{\infty} e^{i\nu\tau}\Phi(L,\nu)d\nu, \quad (32)$$

and similarly the reverse-correlation amplitude is

$$\langle \hat{N}_s(L,\tau)\hat{N}_a(L)\rangle = e^{i\Delta kL}\frac{2\pi}{AN}\int_{-\infty}^{\infty} e^{-i\nu\tau}\Psi(L,\nu)d\nu, \quad (33)$$

with the phase mismatch Δk. The self-correlation amplitudes due to the noise terms are

$$\mathcal{I}_s^n(L,\tau) = \langle \hat{N}_s^\dagger(L,\tau)\hat{N}_s(L)\rangle = \frac{2\pi}{AN}\int_{-\infty}^{\infty} e^{i\nu\tau}S(L,\nu)d\nu, \quad (34)$$

$$\mathcal{I}_a^n(L,\tau) = \langle \hat{N}_a^\dagger(L,\tau)\hat{N}_a(L)\rangle = \frac{2\pi}{AN}\int_{-\infty}^{\infty} e^{-i\nu\tau}A(L,\nu)d\nu, \quad (35)$$

with the spectral functions of the noise products in above equations defined as

$$\Phi(L,\nu) = \sum_{x,x'}\int_0^L 2\tilde{D}_{x,x'}^n(z)C_x^{a*}(\xi,\nu)C_{x'}^s(\xi,\nu)dz, \quad (36)$$

$$\Psi(L,\nu) = \sum_{x,x'}\int_0^L 2\tilde{D}_{x,x'}^{an}(z)C_x^s(\xi,\nu)C_{x'}^{a*}(\xi,\nu)dz, \quad (37)$$

$$S(L,\nu) = \sum_{x,x'}\int_0^L 2\tilde{D}_{x,x'}^n(z)C_x^{s*}(\xi,\nu)C_{x'}^s(\xi,\nu)dz, \quad (38)$$

$$A(L,\nu) = \sum_{x,x'}\int_0^L 2\tilde{D}_{x,x'}^{an}(z)C_x^a(\xi,\nu)C_{x'}^{a*}(\xi,\nu)dz, \quad (39)$$

where $x,x' = ac, ad, bc, bd$ and $\tilde{D}_{x,x'}^{n(an)}(z)$ are the normal (antinormal) ordered diffusion coefficients defined in Appendix E and the coefficients are defined as

$$C_x^s(\xi,\nu) = \frac{1}{g_s}[\bar{U}_s^s(\xi,\nu)X_x(\nu) + \bar{U}_a^s(\xi,\nu)Y_x(\nu)], \quad (40)$$

$$C_x^a(\xi,\nu) = \frac{1}{g_a^*}[\bar{U}_a^a(\xi,\nu)Y_x(\nu) + \bar{U}_s^a(\xi,\nu)X_x(\nu)], \quad (41)$$

with X_x and Y_x given by Eqs. (B9) and (B10).

The adjoint of Eq. (32) can also be expressed in an alternative form $\frac{2\pi e^{i\Delta kL}}{AN}\Sigma_{x,x'}\int_0^L 2\tilde{D}^n_{x,x'}(z)\int_{-\infty}^{\infty}\tilde{C}_x^{a*}(\xi,t')\tilde{C}_{x'}^s(\xi,\tau-t')dt'dz$ using the convolution theorem. If the spatial variations of the populations and the coherences are much slower than $\mathcal{G}_f$ and $\mathcal{K}_f$, the diffusion coefficients $\tilde{D}^{n(an)}_{x,x'}$ can be separated from the functions C_x^s and C_x^a and Eqs. (36)–(39) can be integrated analytically.

B. Boundary products

Analytical expressions for the products of boundary operators can be obtained by using the commutation relation derived in Appendix D $[\hat{E}_f(0,\nu),\hat{E}_f^\dagger(0,\nu')] \simeq C_f\delta(\nu-\nu')$ with $C_f = \frac{\hbar\nu_f\pi}{\varepsilon_0 Ac}$,

$$\langle\hat{B}_a(L,t+\tau)\hat{B}_s(L,t)\rangle = \int\left[C_a(\bar{n}_a+1)\frac{g_a^*}{g_s}\psi_a^{a*}(L,\nu)\psi_a^s(L,\nu) + C_s\bar{n}_s\frac{g_s^*}{g_a}\psi_s^{a*}(L,\nu)\psi_s^s(L,\nu)\right]e^{i\nu\tau}\frac{d\nu}{2\pi}, \quad (42)$$

$$\langle\hat{B}_s(L,t+\tau)\hat{B}_a(L,t)\rangle = \int\left[C_a\bar{n}_a\frac{g_a^*}{g_s}\psi_a^{a*}(L,\nu)\psi_a^s(L,\nu) + C_s(\bar{n}_s+1)\frac{g_s^*}{g_a}\psi_s^{a*}(L,\nu)\psi_s^s(L,\nu)\right]e^{-i\nu\tau}\frac{d\nu}{2\pi}, \quad (43)$$

$$\mathcal{I}_s^b(L,\tau) = \langle\hat{B}_s^\dagger(L,t+\tau)\hat{B}_s(L,t)\rangle = \int_{-\infty}^{\infty}\left[C_a(\bar{n}_a+1)\left|\frac{g_a^*}{g_s}\psi_a^s(L,\nu)\right|^2 + C_s\bar{n}_s|\psi_s^s(L,\nu)|^2\right]e^{i\nu\tau}\frac{d\nu}{2\pi}, \quad (44)$$

$$\mathcal{I}_a^b(L,\tau) = \langle\hat{B}_a^\dagger(L,t+\tau)\hat{B}_a(L,t)\rangle = \int_{-\infty}^{\infty}\left[C_s(\bar{n}_s+1)\left|\frac{g_s}{g_a^*}\psi_s^a(L,\nu)\right|^2 + C_a\bar{n}_a|\psi_a^a(L,\nu)|^2\right]e^{-i\nu\tau}\frac{d\nu}{2\pi}, \quad (45)$$

where $\bar{n}_f = (e^{\beta\hbar\nu_f}-1)^{-1}$ are the mean photon numbers for $f = s,a$.

Equations (42)–(45) give the parts of the correlations in terms of the coefficients of the boundary operators $\hat{E}_s(0,\nu)$ and $\hat{E}_a(0,\nu)$.

The spatially dependent Stokes and anti-Stokes intensities $\mathcal{I}_f^n(L) = \langle\hat{N}_f^\dagger(L,t)\hat{N}_f(L,t)\rangle$ and $\mathcal{I}_f^b(L) = \langle\hat{B}_f^\dagger(L,t)\hat{B}_f(L,t)\rangle$ are obtained from the correlated intensities, Eqs. (34), (35), (44), and (45), by setting $\tau=0$ and will be used to compute the normalized correlations defined in the following section.

Thus, Eqs. (28)–(31) together with the analytical expressions (36)–(39) and (42)–(45) constitute the main results of this paper. Note that the correlations depend only on the relative time delay τ and are independent of the absolute time t even though the solutions $\hat{E}_f(L,t)$ depend on t. Since X_x and Y_x are proportional to N, the correlations are proportional to N^2, showing a collective effect.

V. DEGREE OF NONCLASSICALNESS AND QUANTITATIVE JOINT DETECTION RATE

The finite value of Glauber's two-photon correlation $G^{(2)}$ does not necessarily imply that the correlation is quantum mechanical or nonclassical. Let us recall that $g^{(2)}(0) < g^{(2)}(\tau)$ (antibunching) and $g^{(2)}(\tau) < 1$ (sub-Poissonian) correspond to nonclassical correlation, with $g^{(2)}=1$ for the coherent state and $g^{(2)}=2$ for the thermal state. The photon statistics of the Stokes and anti-Stokes photons can be determined from the normalized self-correlations [23]

$$g_{ff}^{(2)}(L,\tau) \doteq G_{ff}^{(2)}(L,\tau)/\mathcal{I}_f(L)^2. \quad (46)$$

The existence of a second order of coherence $g_{as}^{(2)}=1$ and the degree of correlation $(g_{as}^{(2)}-1)$ between the Stokes and anti-Stokes fields can be seen from the normalized cross (reverse) correlation

$$g_{as(sa)}^{(2)}(L,\tau) \doteq G_{as(sa)}^{(2)}(L,\tau)/\mathcal{I}_s(L)\mathcal{I}_a(L). \quad (47)$$

The degree of nonclassical correlation can be quantified by defining the Cauchy-Schwartz cross (reverse) correlation

$$g_{as(sa)}^{CS}(L,\tau) = G_{as(sa)}^{(2)}(L,\tau)/\sqrt{G_{ss}^{(2)}(L,\tau)G_{aa}^{(2)}(L,\tau)}, \quad (48)$$

where the nonclassical regime $g_{as}^{CS}>1$ corresponds to violation of the Cauchy-Schwartz inequality. A larger value of g_{as}^{CS} indicates a large degree of nonclassical correlation. This is justified, for example, in the case of sub-Poissonian correlation $[g_{ss}^{(2)}(0)<1]$ and large cross correlation $g_{as}^{(2)}>>1$.

We now relate our results for the two-photon correlation with the experimental detection rate. The detection rate of Stokes or anti-Stokes photons can be expressed as

$$R_f = \frac{n_f}{t_{\rm det}} = \frac{4\mathcal{I}_f 2\varepsilon_0 V_{\rm det}}{\hbar\nu_f t_{\rm det}} = 8\varepsilon_0\mathcal{I}_f A_{\rm det}c/\hbar\nu_f, \quad (49)$$

where $A_{\rm det}=\frac{1}{4}\pi d^2$ is the area of the detector and we have used $V_{\rm det}=A_{\rm det}z_{\rm det}$ and $c=z_{\rm det}/t_{\rm det}$. The Stokes and anti-Stokes

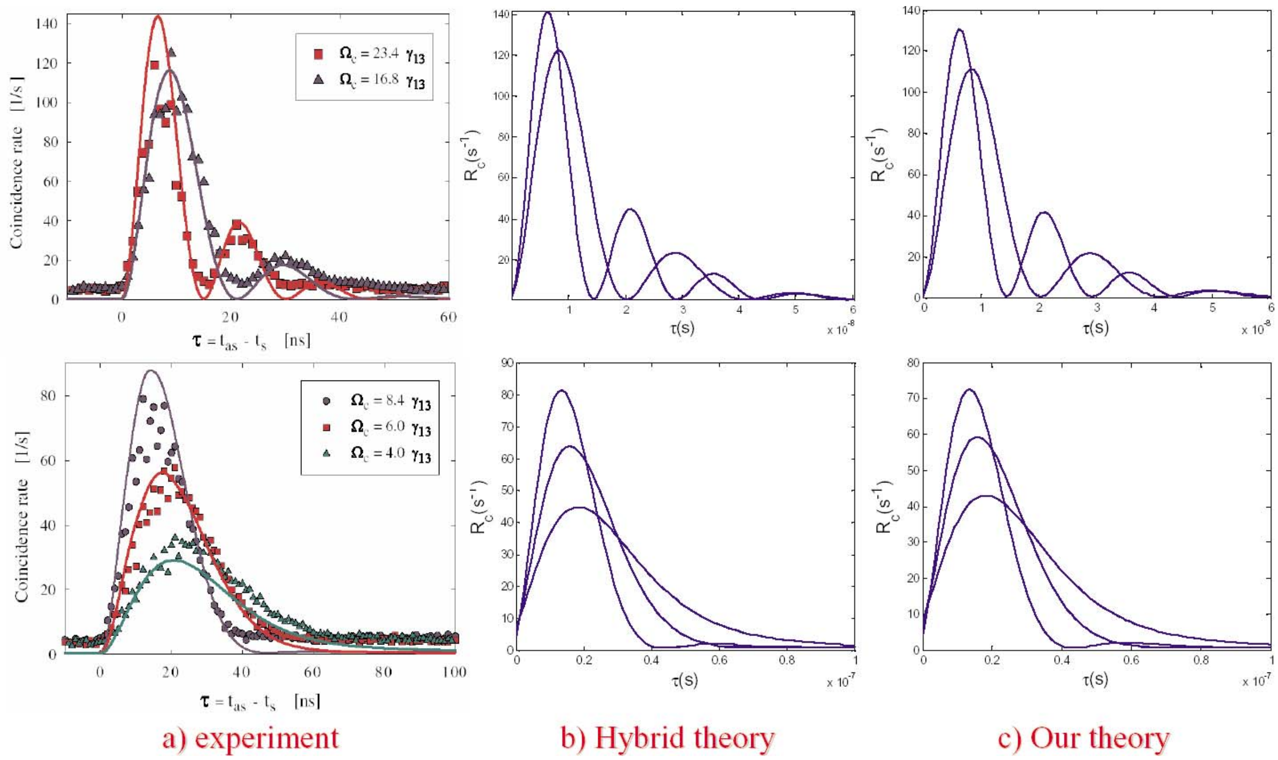

FIG. 2. (Color online) Comparison of the joint detection results computed from the cross correlation, Eqs. (28) and (51). (a) Experimental results of Ref. [9]. (b) Hybrid theory based on the coupled equations of Balic *et al.* [9] and our analytical approach to calculate $G^{(2)}$. Quantitative agreement is obtained when the coefficients ξ_s and ξ_{as} in [9] are multiplied by $\sqrt{N\sigma_{db}}$ and σ is replaced by σ_{ac} as in Ref. [25] to obtain the correct units. (c) Our complete theory. The curves in (b) and (c) are plotted using the experimental parameters of the decay rates and dipole moments [26] for ^{87}Rb with optical depth $N\sigma_{ac}L=2(g_a\kappa_a)L/\gamma_{ac}=11$ (where we define the cross sections $\sigma_{ac}=\frac{|\wp|^2\omega_{ac}}{\hbar c\varepsilon_o\gamma_{ac}}$, $\sigma_{db}=\frac{|\wp|^2\omega_{db}}{\hbar c\varepsilon_o\gamma_{db}}$), weak pump $\Omega_p=0.8\gamma_{ac}/2$, detuning $\Delta=-7.5\gamma_{ac}$, decoherence $\gamma_{bc}=0.6\gamma_{ac}$, $\Omega_c/\gamma_{ac}=23.4/2$, 16.8/2, 8.4/2, 6/2, and 4/2. The factor of 1/2 is due to a different definition of the Rabi frequency in our case. Quantitative agreement is obtained by multiplying R_c by 16 due to the fact that the electric field defined is twice the physical field. We take the effective transverse cross section as $A=(\pi w_o^2)\sqrt{3/4}$ where $w_o=w_1/\sqrt{2}$ is the waist and $2w_1=280$ μm is the diameter at $1/e^2$ intensity [9] of the pump laser. The factor of $\sqrt{3/4}$ leads to a smaller effective area by taking into account the transverse variation of the laser beam.

wavelengths are $\lambda_s=780$ nm and $\lambda_a=795$ nm. According to [9], the correlated detection rate [as reproduced in Fig. 2(a)] is related to the normalized correlation as

$$R_c = \bar{G}^{(2)}\epsilon^2\Delta T = g_{as}^{(2)}R_sR_a\epsilon^2\Delta T, \tag{50}$$

where $\bar{G}^{(2)}$ is an integrated correlation and has units of s^{-2}, ϵ is the detection efficiency, and ΔT is the bin width. From $g_{as}^{(2)} \doteq \frac{G_{as}^{(2)}}{\mathcal{I}_s\mathcal{I}_a}$ we have a useful relation between the correlated detection rate and Glauber's absolute correlation $G_{as}^{(2)}$:

$$R_c = \frac{G_{as}^{(2)}}{\mathcal{I}_s\mathcal{I}_a}R_sR_a\epsilon^2\Delta T = \frac{G_{as}^{(2)}}{\nu_a\nu_s}\left(\frac{8\epsilon\varepsilon_0A_{\rm det}c}{\hbar}\right)^2\Delta T. \tag{51}$$

Using the values of Ref. [9], $d=5.6$ μm (for single-mode fiber), $\epsilon=0.3$, and $\Delta T=1$ ns in Eq. (51) and using Eq. (28) (including boundary operators), we compute the theoretical rates shown in Fig. 2(b) using the coupled equations of Balic *et al.* and Fig. 2(c) using our coupled equations. We obtain good quantitative agreement with the experimental data [9] reproduced in Fig. 2(a). We find that results with the boundary operators alone do not give good quantitative agreement with experimental data. This also shows that both the boundary operators and the noise operators are necessary to provide a correct quantitative description of the two-photon correlation. The comparison between noise and boundary operators will be elaborated on in a subsequent paper.

VI. DISCUSSIONS AND CONCLUSIONS

We have presented a complete analytical quantum theory of the two-photon correlations $G_{as}^{(2)}$, $G_{sa}^{(2)}$, and $G_{ff}^{(2)}$ for the macroscopic Stokes and anti-Stokes fields generated in an amplifier (extended coherent medium) in a double-Λ scheme. We have derived coupled parametric oscillator equations valid for arbitrary detuning and intensity of the lasers and analytical expressions for the correlations. The hybrid results [Fig. 2(b)], obtained by using our analytical expression of the correlations along with the coupled equations of Balic *et al.* [9] for the RED scheme give good quantitative agreement with the experimental data [Fig. 2(a)] without any fitting parameter for both high- and low-control Ω_c fields. Similarly, the results obtained using our more general coupled equations [Fig. 2(c)] correspond quite well with the hybrid results.

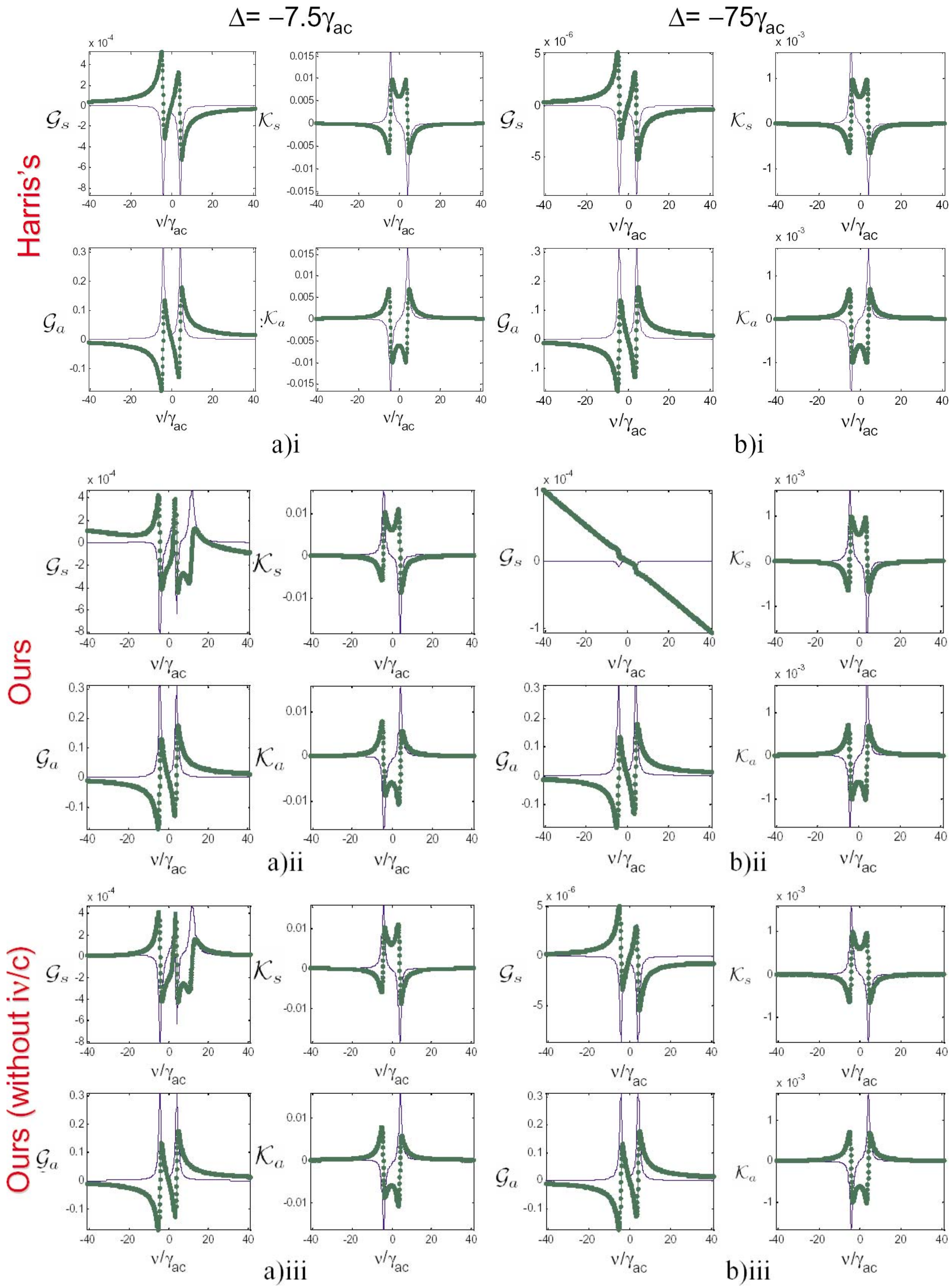

FIG. 3. (Color online) Comparison of the real (solid line) and imaginary (dotted line) pairs of the gain and loss $\mathcal{G}_{s,a}(\nu)$ and coupling $\mathcal{K}_{s,a}(\nu)$ coefficients for (a) $\Delta=-7.5\gamma_{ac}$ (as used in experiment) and (b) $\Delta=-75\gamma_{ac}$ (larger detuning). Other parameters follow from the experiment [9]: $\Omega_p=0.8\gamma_{ac}/2$, $\Omega_c=8.4\gamma_{ac}/2$, and $N\sigma_{ac}L=11$. Cases (i) coefficients of Balic *et al.* [9], (ii) our coefficients, Eqs. (7) and (8), and (iii) our coefficients but $i\nu/c$ is neglected.

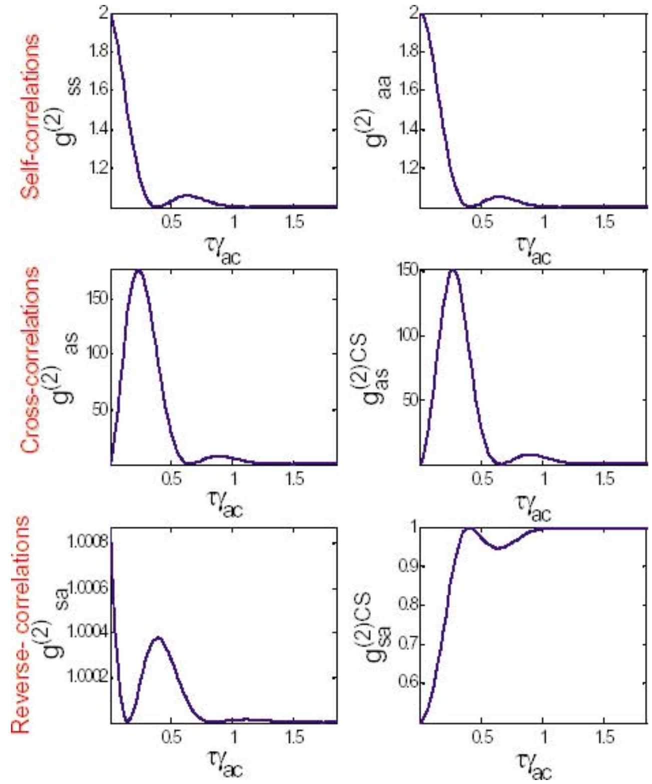

FIG. 4. (Color online) Normalized self-correlations $g_{ff}^{(2)n}$, cross correlation $g_{as}^{(2)n}$, and reverse correlation $g_{sa}^{(2)n}$ with Cauchy-Schwartz cross correlation g_{sa}^{CS} and reverse correlation g_{as}^{CS} for the experimental parameters $N\sigma L=11$, $\Omega_p=0.8\gamma_{ac}/2$, $\Delta=-7.5\gamma_{ac}$, $\gamma_{bc}=0.6\gamma_{ac}$, and $\Omega_c/\gamma_{ac}=8.4/2$.

The above correspondence can be understood through the following analysis. Figure 3(aii) shows that all our coefficients ($\mathcal{G}_a$, $\mathcal{K}_a$, and $\mathcal{K}_s$) except $\mathcal{G}_s$ agree almost perfectly with the coefficients of Balic *et al.* given in Eqs. (B11)–(B14). By $\mathcal{G}_s$ from Eq. (7) with $w_{bb}^{st}\simeq\tilde{\rho}_{ba}^{st}\simeq 0$ and $\tilde{\rho}_{cd}^{st}\simeq -\frac{1}{T_{dc}}i\Omega_p^*$, then applying the conditions for the RED scheme, $[\frac{T_{ad}(\nu)}{T_{bc}(\nu)}>>1$, $T_{ac}(\nu)>>\frac{|\Omega_p|^2}{T_{ad}(\nu)}$, $T_{db}^*(\nu)>>\frac{|\Omega_c|^2}{T_{ad}(\nu)}]$ and neglecting the term with $i\frac{\nu}{c}$, we obtain

$$\mathcal{G}_s\simeq -\frac{|\Omega_p|^2}{T_{dc}T_{db}^*(\nu)}\frac{g_s\kappa_sT_{ac}(\nu)}{T_{bc}(\nu)T_{ac}(\nu)+|\Omega_c|^2}, \tag{52}$$

which reduces exactly to the expression of Balic *et al.*, Eq. (B11) if we use $\Delta>>\gamma_{dc},\gamma_{db}$. Thus, our analytical results are consistent with the theory of Balic *et al.* even the though our coupled equations depend on additional parameters not found in the coupled equations of Balic *et al.* [12]—i.e., the decoherence rates γ_{dc}, γ_{db}, and γ_{ab}.

The asymmetric feature of our $\mathcal{G}_s$ [Figs. 3(aii) and 3(aiii)] across $\nu=0$ is due to the quasiresonant effect of the pump and the coexistence with the control field. For larger detuning, Fig. 3(biii) shows that $\mathcal{G}_s$ becomes symmetric and agrees very well with the coefficient of Balic *et al.*, Fig. 3(bi), while we confirm that the corresponding correlations agree perfectly. The divergence of the imaginary component of our $\mathcal{G}_s$ as $|\nu|$ increases is due to $i\frac{\nu}{c}$ corresponding to the time derivative. This can be verified by comparing with the case when $i\frac{\nu}{c}$ is neglected: namely, Figs. 3(aii) and 3(aiii) or 3(bii) and 3(biii). However, $i\frac{\nu}{c}$ is an odd function which integrates to give zero (when $\beta_{s,a}<<1$) and therefore does not affect the results. If the detunings and Rabi frequencies of the two lasers are the same, we have $\mathcal{G}_s=\mathcal{G}_a$ and $\mathcal{K}_s=\mathcal{K}_a$. To summarize, the correctness of our generalized coefficients have been verified and they include the coexistence of the pump and control fields and the coherence between upper levels a and d, which give rise to additional resonant features in the spectra, particularly of $\mathcal{G}_s$.

In Fig. 4, we plot the normalized self-correlations, cross correlation, reverse correlation, and Cauchy-Schwartz correlation defined in Sec. V. The self-correlations change from thermal ($g_{ff}^{(2)}=2$) to coherent ($g_{ff}^{(2)}=1$) nature as the time delay increases, but remain classical. The cross correlation shows a large nonclassical correlation ($g_{as}^{(2)}$, $g_{as}^{CS}>>1$). The reverse correlation is negligible [15] ($g_{sa}^{(2)}\sim 1$) and remains classical $g_{sa}^{CS}<1$ which is consistent with bunching in $g_{sa}^{(2)}$.

Finally, we conclude that the correspondence of our generalized coupled equations and the ability of our analytical expression for the correlation to provide good quantitative agreement with experimental data allow us to forecast new results in different regimes of laser parameters in a series of subsequent papers.

ACKNOWLEDGMENTS

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APPENDIX A: CLOSED SET OF HEISENBERG-LANGEVIN-MAXWELL EQUATIONS

The Hamiltonian for the four-level system [Fig. 1(a)] in the Schrödinger picture is $H=\Sigma_j\{H_{0j}+V_j\}$ where

$$H_{0j}=\sum_{\mathrm{s}=a,b,c,d}\hbar\omega_{\mathrm{s}}|\mathrm{s}^j\rangle\langle\mathrm{s}^j|+\sum_{\mathbf{k},\lambda}\left(\hat{a}_{\mathbf{k}\lambda}^{\dagger}\hat{a}_{\mathbf{k}\lambda}+\frac{1}{2}\right)\hbar\nu_{\mathbf{k}\lambda} \tag{A1}$$

and

$$V_j=-\hbar\Bigg[\sum_{\mathbf{k},\mathrm{s}=b,c}(g_{\mathbf{k}}^{a\mathrm{s}}|a^j\rangle\langle\mathrm{s}^j|+g_{\mathbf{k}}^{d\mathrm{s}}|d^j\rangle\langle\mathrm{s}^j|)\hat{a}_{\mathbf{k}}(t)e^{i\mathbf{k}\cdot\mathbf{r}_j}+\Omega_p|d^j\rangle \times\langle c^j|e^{i(\mathbf{k}_p\cdot\mathbf{r}_j-\nu_pt)}+\Omega_c|a^j\rangle\langle b^j|e^{i(\mathbf{k}_c\cdot\mathbf{r}_j-\nu_ct)}+g_s\hat{E}_s|d^j\rangle \times\langle b^j|e^{i(\mathbf{k}_s\cdot\mathbf{r}_j-\nu_st)}+g_a\hat{E}_a|a^j\rangle\langle c^j|e^{i(\mathbf{k}_a\cdot\mathbf{r}_j-\nu_at)}+\mathrm{adj}\Bigg] \tag{A2}$$

are the free Hamiltonian and interaction Hamiltonian in the Schrödinger picture for single particles in the medium and $g_s=\wp_{db}/\hbar$, $g_a=\wp_{ac}/\hbar$, $g_{\mathbf{k}}^{a\mathrm{s}}=\wp_{a\mathrm{s}}\mathcal{E}_{\mathbf{k}}/\hbar$, and $\mathcal{E}_{\mathbf{k}}=\sqrt{\hbar\nu_{\mathbf{k}}/2\varepsilon_o\mathcal{V}}$.

The subscripts p, s, q, and a stand for pump, Stokes control and anti-Stokes fields, respectively. The number density N in the extended medium of volume V is assumed to be sufficiently large $n=VN>>1$ such that the discrete operators can be converted into continuous variables operators:

$$\frac{(2\pi)^2}{AN}\sum_{j=1}^{n}\hat{\sigma}^j_{\beta\alpha}(t)\delta(z-z_j)\rightarrow\hat{\sigma}_{\beta\alpha}(z,t), \tag{A3}$$

$$e^{-i\mathbf{k}_{\beta\alpha}\cdot\mathbf{r}_j}\rightarrow e^{-ik_{\beta\alpha}z}, \tag{A4}$$

$$\frac{(2\pi)^2}{AN}\sum_{j=1}^{n}\hat{F}^j_{\beta\alpha}(t)\delta(z-z_j)\rightarrow\hat{F}_{\beta\alpha}(z,t), \tag{A5}$$

where $\hat{\sigma}_{\beta\alpha}(t)=|\beta^j\rangle\langle\alpha^j|$, $\hat{F}^j_{\beta\alpha}(t)$ are the quantum noise operators, and A is the effective transverse interaction area covered by the laser beams. However, the interparticle distance $d \sim N^{-1/3}$ is larger than the optical wavelength λ so that the dipole-dipole interaction can be neglected. Hence, we obtain a set of 16 Heisenberg-Langevin atomic operator equations in continuous variables coupled to the 4 (including the adjoints) propagation equations for the Stokes $\hat{E}_s$ and anti-Stokes $\hat{E}_s$.

The above equations can be simplified to a closed set of equations that can be solved exactly for the field operators $\hat{E}_{s,a}$ if $\hat{p}_{xx}$ and $\hat{p}_{ab},\hat{p}_{cd}$ are taken as c numbers, corresponding to the steady-state density matrix elements. The coupled equations are

$$\frac{d}{dt}\hat{p}_{ac}=-T_{ac}\hat{p}_{ac}-ig_a^*\hat{E}_a^\dagger(\hat{p}_{cc}-\hat{p}_{aa})-i(\Omega_c^*\hat{p}_{bc}-\Omega_p^*\hat{p}_{ad}) + e^{ik_az}e^{-i\nu_at}\hat{F}_{ac}, \tag{A6}$$

$$\frac{d}{dt}\hat{p}_{ad}=-T_{ad}\hat{p}_{ad}+i(g_s\hat{p}_{ab}\hat{E}_s-g_a^*\hat{E}_a^\dagger\hat{p}_{cd})+i(\Omega_p\hat{p}_{ac}-\Omega_c^*\hat{p}_{bd}) + e^{ik_{qs}z}e^{-i\nu_{qs}t}\hat{F}_{ad}, \tag{A7}$$

$$\frac{d}{dt}\hat{p}_{bc}=-T_{bc}\hat{p}_{bc}-i(g_s\hat{p}_{dc}\hat{E}_s-g_a^*\hat{E}_a^\dagger\hat{p}_{ba})-i(\Omega_c\hat{p}_{ac}-\Omega_p^*\hat{p}_{bd}) + e^{-ik_{qa}z}e^{i\nu_{qa}t}\hat{F}_{bc}, \tag{A8}$$

$$\frac{d}{dt}\hat{p}_{bd}=-T_{db}^*\hat{p}_{bd}+ig_s(\hat{p}_{bb}-\hat{p}_{dd})\hat{E}_s+i(\Omega_p\hat{p}_{bc}-\Omega_c\hat{p}_{ad}) + e^{-ik_sz}e^{i\nu_st}\hat{F}_{db}^\dagger, \tag{A9}$$

$$\left(\frac{1}{c}\frac{\partial}{\partial t}+\frac{\partial}{\partial z}\right)\hat{E}_s(z,t)=i\kappa_s\hat{p}_{bd}(z,t), \tag{A10}$$

$$\left(\frac{1}{c}\frac{\partial}{\partial t}\pm\frac{\partial}{\partial z}\right)\hat{E}_a^\dagger(z,t)=-i\kappa_a^*\hat{p}_{ac}(z,t), \tag{A11}$$

with $\hat{F}_{db}^\dagger=\hat{F}_{bd}$ and the slowly varying atomic operators $\hat{p}_{ac}=\hat{\sigma}_{ac}e^{ik_az}e^{-i\nu_at}$, $\hat{p}_{ad}=\hat{\sigma}_{ad}e^{ik_{qs}t}e^{-i\nu_{qs}t}$, $\hat{p}_{bc}=\hat{\sigma}_{bc}e^{ik_{aq}z}e^{-i\nu_{aq}t}$, and $\hat{p}_{bd}=\hat{\sigma}_{bd}e^{-ik_st}e^{i\nu_st}$. The "−" is for counterpropagating geometry, and the complex decoherences are

$$T_{ac}=i(\nu_a-\omega_{ac})+\gamma_{ac}, \tag{A12}$$

$$T_{ad}=i(\nu_c-\nu_s-\omega_{ad})+\gamma_{ad}, \tag{A13}$$

$$T_{bc}=i(\nu_p-\nu_s-\omega_{bc})+\gamma_{bc}, \tag{A14}$$

$$T_{db}=i(\nu_s-\omega_{db})+\gamma_{db}, \tag{A15}$$

with the decoherence rates

$$\gamma_{ac}\doteq\frac{1}{2}\{\Gamma_{ac}(2\bar{n}_{ac}+1)+\Gamma_{ab}(\bar{n}_{ab}+1)\}+\gamma_{ac}^{dep}, \tag{A16}$$

$$\gamma_{ad}=\frac{1}{2}\{\Gamma_{db}(\bar{n}_{db}+1)+\Gamma_{dc}(\bar{n}_{dc}+1)+\Gamma_{ab}(\bar{n}_{ab}+1) + \Gamma_{ac}(\bar{n}_{ac}+1)\}+\gamma_{ad}^{dep}, \tag{A17}$$

$$\gamma_{bc}=\frac{1}{2}\{\Gamma_{ab}\bar{n}_{ab}+\Gamma_{db}\bar{n}_{db}+\Gamma_{ac}\bar{n}_{ac}+\Gamma_{dc}\bar{n}_{dc}\}+\gamma_{bc}^{dep}, \tag{A18}$$

$$\gamma_{db}\doteq\frac{1}{2}\{\Gamma_{db}(2\bar{n}_{db}+1)+\Gamma_{dc}(\bar{n}_{dc}+1)\}+\gamma_{db}^{dep}, \tag{A19}$$

where $\Gamma_{\alpha\beta}$ are the spontaneous emission rates and $\bar{n}_f=(e^{\beta\hbar\nu_f}-1)^{-1}$ and $\gamma_{\alpha\beta}^{dep}$ are the dephasings due to phonons in the condensed phase or atomic collisions in gas. Here, we consider a cold sample with negligible inhomogeneous broadening $(1/T_2^*\rightarrow 0)$.

APPENDIX B: COEFFICIENTS FOR COUPLED PARAMETRIC EQUATIONS

We focus on the quasistatic regime which enables us to solve either Eqs. (1) and (2) or Eqs. (5) and (6) by Fourier transforming the t variable to ν. The resulting equations can then be solved algebraically by Laplace transforming the z variable to q. The inverse Laplace transform gives the solutions (10) and (11). The gain and loss coefficients are

$$G_s=-\alpha_s\left\{w_{bb}^{st}+\frac{i\Omega_c}{T_{ad}(\nu)}\tilde{\rho}_{ba}^{st}+\frac{i\Omega_p}{T_{bc}(\nu)}\tilde{\rho}_{cd}^{st}\right\}-i\frac{\nu}{c}, \tag{B1}$$

$$G_a=-\alpha_a\left\{w_{cc}^{st}-\frac{i\Omega_c^*}{T_{bc}(\nu)}\tilde{\rho}_{ab}^{st}-\frac{i\Omega_p^*}{T_{ad}(\nu)}\tilde{\rho}_{dc}^{st}\right\}-i\frac{\nu}{c}, \tag{B2}$$

where the superscript "st" implies steady state and $T_x(\nu)=T_x-i\nu$. The $\tilde{\rho}_{\beta\alpha}^{st}=\langle\hat{p}_{\alpha\beta}^{st}(\infty)\rangle$ and inversions $w_{cc}^{st}=\rho_{aa}^{st}-\rho_{cc}^{st}$, $w_{bb}^{st}=\rho_{dd}^{st}-\rho_{bb}^{st}$ are the steady-state solutions of the density matrix equations. Note that the imaginary parts of $\tilde{\rho}_{ba}^{st}$ and $\tilde{\rho}_{cd}^{st}$ lead to amplification $\mathrm{Re}\{G_s,G_a\}<0$ even without inversion $w_{bb}^{st},w_{cc}^{st}<0$.

The term $i\frac{\nu}{c}$ is due to the time derivatives of the field operators. It will not affect the final result for the correlation.

This has been verified numerically and can be understood analytically if we use Eqs. (A6)–(A11) in the retarded frame through the new variables $t'=t-z/c$.

The cross couplings are

$$K_s = i\alpha_s\left\{\frac{\Omega_p}{T_{bc}(\nu)}\tilde{\rho}_{ab}^{st} + \frac{\Omega_c}{T_{ad}(\nu)}\tilde{\rho}_{dc}^{st}\right\} - i\frac{\nu}{c}\beta_s, \qquad (B3)$$

$$K_a = -i\alpha_a\left\{\frac{\Omega_p^*}{T_{ad}(\nu)}\tilde{\rho}_{ba}^{st} + \frac{\Omega_c^*}{T_{bc}(\nu)}\tilde{\rho}_{cd}^{st}\right\} - i\frac{\nu}{c}\beta_a, \qquad (B4)$$

with the effective propagation coefficients

$$\alpha_s = \frac{g_s\kappa_s}{T_{db}^*(\nu)+\delta_s}, \quad \alpha_a = \frac{g_a^*\kappa_a^*}{T_{ac}(\nu)+\delta_a} \qquad (B5)$$

and the power broadening frequencies

$$\delta_s = \frac{|\Omega_c|^2}{T_{ad}(\nu)} + \frac{|\Omega_p|^2}{T_{bc}(\nu)}, \quad \delta_a = \frac{|\Omega_p|^2}{T_{ad}(\nu)} + \frac{|\Omega_c|^2}{T_{bc}(\nu)}, \qquad (B6)$$

and the self-coupling coefficients appearing in Eqs. (5) and (6) are

$$\beta_s = \frac{g_s\kappa_s}{g_a^*\kappa_a^*}\frac{\Omega_c\Omega_p}{T_{db}^*(\nu)+\delta_s}\left(\frac{1}{T_{bc}(\nu)} + \frac{1}{T_{ad}(\nu)}\right), \qquad (B7)$$

$$\beta_a = \frac{g_a^*\kappa_a^*}{g_s\kappa_s}\frac{\Omega_c^*\Omega_p^*}{T_{ac}(\nu)+\delta_a}\left(\frac{1}{T_{bc}(\nu)} + \frac{1}{T_{ad}(\nu)}\right), \qquad (B8)$$

with the propagation constants $\kappa_s = N\wp_{bd}c\mu_o\nu_s/2$ and $\kappa_a = N\wp_{ca}c\mu_o\nu_a/2$. The real parts of $G_{s,a}$ are even functions across ν and the imaginary parts are odd functions and vice-versa for $K_{s,a}$.

The coefficients in Eqs. (3) and (4) are

$$X_{ad} = \alpha_s\frac{\Omega_c}{T_{ad}(\nu)}, \quad X_{bc} = -\alpha_s\frac{\Omega_p}{T_{bc}(\nu)}, \quad X_{bd} = i\alpha_s, \qquad (B9)$$

$$Y_{ad} = \alpha_a\frac{\Omega_p^*}{T_{ad}(\nu)}, \quad Y_{bc} = -\alpha_a\frac{\Omega_c^*}{T_{bc}(\nu)}, \quad Y_{ac} = -i\alpha_a. \qquad (B10)$$

For comparison with our coefficients, we rewrite the coefficients of Balic *et al.* [9] (Superscript B) based on a more precise form of Ref. [25] in terms of our notation $\Omega_c = \Omega_c^B/2$:

$$\mathcal{G}_s = -T_{ac}(\nu)\left(\frac{\Omega_p}{\Delta}\right)^2\frac{\frac{1}{2}N\sigma_{db}\gamma_{db}}{D^*(\nu)}, \qquad (B11)$$

$$\mathcal{G}_a = T_{bc}(\nu)\frac{\frac{1}{2}N\sigma_{ac}\gamma_{ac}}{D^*(\nu)}, \qquad (B12)$$

$$\mathcal{K}_s = -i\frac{\Omega_p^*\Omega_c^*}{\Delta}\frac{\frac{1}{2}N\sigma_{db}\gamma_{db}}{D^*(\nu)}, \qquad (B13)$$

$$\mathcal{K}_a = i\frac{\Omega_p^*\Omega_c^*}{\Delta}\frac{\frac{1}{2}N\sigma_{ac}\gamma_{ac}}{D^*(\nu)}, \qquad (B14)$$

$$X_{bc} = -iT_{ac}(\nu)\frac{\Omega_p^*}{\Delta}\frac{N\sqrt{\sigma_{db}\sigma_{ac}\gamma_{db}\gamma_{ac}}}{D^*}, \qquad (B15)$$

$$X_{ac} = -\Omega_c^*\left(\frac{\Omega_p}{\Delta}\right)^2\frac{N\sigma_{ac}\gamma_{ac}}{D^*}, \qquad (B16)$$

$$Y_{bc} = -\Omega_c^*\frac{N\sqrt{\sigma_{db}\sigma_{ac}\gamma_{db}\gamma_{ac}}}{D^*}, \qquad (B17)$$

$$X_{ac} = -iT_{bc}(\nu)\frac{N\sigma_{ac}\gamma_{ac}}{D}, \qquad (B18)$$

where $D(\nu) = |\Omega_c|^2 - (i\gamma_{ac}-\nu)(i\gamma_{bc}-\nu) = D^B/4$ with $N\sigma_{db}\gamma_{db} = 2g_s\kappa_s$, $N\sigma_{ac}\gamma_{ac} = 2g_a\kappa_a$, and $\sigma_{db} = \frac{|\wp|^2\omega_{db}}{\hbar c\varepsilon_o\gamma_{db}}$, $\sigma_{ac} = \frac{|\wp|^2\omega_{ac}}{\hbar c\varepsilon_o\gamma_{ac}}$. Note that our definition of the Fourier transform is opposite to that of Balic *et al.* [9].

APPENDIX C: DERIVATION OF Eq. (32)

We derive the cross correlation of the noise terms in Eq. (28) using the solutions in frequency space, Eqs. (10) and (11), as follows:

$$\langle\hat{N}_a(t+\tau)\hat{N}_s(t)\rangle = \int_{-\infty}^{\infty} e^{i\nu_2(t+\tau)}\frac{d\nu_2}{2\pi}\int_{-\infty}^{\infty} e^{-i\nu_1 t}\frac{d\nu_1}{2\pi}\langle\hat{N}_a(z,\nu_2)\hat{N}_s(z,\nu_1)\rangle$$
$$= e^{i\Delta kz}\int_{-\infty}^{\infty} e^{i\nu_2(t+\tau)}\frac{d\nu_2}{2\pi}\int_{-\infty}^{\infty} e^{-i\nu_1 t}\frac{d\nu_1}{2\pi}\sum_{x,x'}\int_0^z dz_2\int_0^z dz_1 C_x^{a*}(\xi_2,\nu_2)C_{x'}^s(\xi_1,\nu_1)\langle\hat{G}_x^\dagger(z_2,\nu_2)\hat{G}_{x'}(z_1,\nu_1)\rangle. \qquad (C1)$$

The noise products in the frequency domain are related to the diffusion coefficients $2D_{x,x'}^n$ defined in Appendix E,

$$\langle \hat{G}_x^\dagger(z_2,\nu_2)\hat{G}_{x'}(z_1,\nu_1)\rangle = \int_{-\infty}^{\infty} dt_2 e^{-i\nu_2 t_2}\int_{-\infty}^{\infty} dt_1 e^{i\nu_1 t_1} e^{-i\theta_x} e^{i\theta_{x'}} \langle \hat{F}_x^\dagger(z_2,t_2)\hat{F}_{x'}(z_1,t_1)\rangle$$
$$= \int_{-\infty}^{\infty} dt_2 e^{-i\nu_2 t_2}\int_{-\infty}^{\infty} dt_1 e^{i\nu_1 t_1}\frac{(2\pi)^2}{AN} 2\tilde{D}^n_{x,x'}(z_1,t_1)\,\delta(t_2-t_1)\,\delta(z_2-z_1), \tag{C2}$$

where $2\tilde{D}^n_{x,x'}(z_1,t_1)=2D^n_{x,x'}e^{-i\theta_x}e^{i\theta_{x'}}$ are the slowly varying diffusion coefficients. Since $2\tilde{D}^n_{x,x'}$ vary much slower than the exponentials $e^{-i\nu_2 t_2}$ and $e^{i\nu_1 t_1}$, we can take their steady state-values $2\tilde{D}^n_{x,x'}(z_1)$ but keep the spatial dependence. We then have a δ function in frequency that enables further simplification of the number of integrals:

$$\langle \hat{G}_x^\dagger(z_2,\nu_2)\hat{G}_{x'}(z_1,\nu_1)\rangle = \frac{(2\pi)^3}{AN} 2\tilde{D}^n_{x,x'}(z_1)\,\delta(z_2-z_1)\,\delta(\nu_1-\nu_2). \tag{C3}$$

Finally, we have

$$\langle \hat{N}_a(t+\tau)\hat{N}_s(t)\rangle = e^{i\Delta k z}\frac{2\pi}{AN}\sum_{x,x'}\int_{-\infty}^{\infty} e^{i\nu\tau} \times \int_0^z 2\tilde{D}^n_{x,x'}(z) C_x^{a*}(\xi,\nu) C^s_{x'}(\xi,\nu)\,dz\,d\nu, \tag{C4}$$

which is identical to the first term of Eq. (32).

APPENDIX D: COMMUTATION RELATION FOR BOUNDARY FIELD OPERATORS

The Stokes and anti-Stokes operators at boundary are

$$\tilde{E}_f(0,t) = \mathcal{E}_f \sum_{\mathbf{k}} \hat{a}_{\mathbf{k}} e^{-i\nu_{\mathbf{k}} t} e^{ik_x x + ik_y y}, \tag{D1}$$

$$\hat{E}_f(0,\nu) = \mathcal{E}_f \sum_{\mathbf{k}} \hat{a}_{\mathbf{k}} 2\pi\delta(\nu-\nu_{\mathbf{k}}) e^{ik_x x + ik_y y}, \tag{D2}$$

where $\mathcal{E}_f=\sqrt{\hbar\nu_f/2\varepsilon_0\mathcal{V}}$. Using $[\hat{a}_{\mathbf{k}},\hat{a}^\dagger_{\mathbf{k}'}]=\delta_{\mathbf{k},\mathbf{k}'}$ we find

$$[\hat{E}_f(0,\nu),\hat{E}_f^\dagger(0,\nu')] = (2\pi\mathcal{E}_f)^2 \sum_{\mathbf{k}} \delta(\nu-\nu_{\mathbf{k}})\,\delta(\nu'-\nu_{\mathbf{k}}). \tag{D3}$$

The conversion to integration using $\Sigma_{\mathbf{k}}\ldots \to \frac{A}{(2\pi)^2}\int_\perp d^2k \frac{L}{2\pi}\int dk \ldots \simeq \frac{L}{2\pi}\int dk\cdots$ gives

$$[\hat{E}_f(0,\nu),\hat{E}_f^\dagger(0,\nu')] = \frac{\hbar\nu_f}{2\varepsilon_0 V}\frac{2\pi L}{c}\int \delta(\nu-u)\,\delta(\nu'-u)\,du$$
$$\simeq \frac{\hbar\nu_f\pi}{\varepsilon_0 A c}\delta(\nu-\nu'), \tag{D4}$$

where we have used the identity $\int f(u)\delta(\nu-u)\delta(\nu'-u)du = f(\nu)\delta(\nu-\nu')$ and $\mathcal{V}=AL$.

APPENDIX E: DIFFUSION COEFFICIENTS

The quantum noise correlation for two discrete particles is δ correlated in time $\langle \hat{F}^j{}_x(t)\hat{F}^k{}_{x'}(t')\rangle = 2D^j_{x,x'}(t)\,\delta(t-t')\,\delta_{jk}$. So, in one spatial dimension,

$$\langle \hat{F}_x(z,t)\hat{F}_{x'}(z',t')\rangle = \frac{(2\pi)^4}{(AN)^2}\sum_{j,k=1}^{n}\langle \hat{F}_x^j(t)\,\delta(z-z_j)\hat{F}^k_{x'}(t') \times \delta(z'-z_k)\rangle$$
$$= \frac{(2\pi)^4}{(AN)^2}\sum_{j=1}^{n} 2D^j_{x,x'}(t)\,\delta(t-t')\,\delta(z-z_j) \times \delta(z'-z_j)$$
$$\simeq \frac{(2\pi)^2}{AN} 2D_{x,x'}(z,t)\,\delta(t-t')\,\delta(z-z'), \tag{E1}$$

where $D_{x,x'}(z,t)=\frac{(2\pi)^2}{AN}\Sigma_{j=1}^n D^j_{x,x'}(t)\,\delta(z-z_j)$.

Hence, the normal-ordered noise correlations are related to the diffusion coefficients as

$$\langle \hat{F}_x^\dagger(z,t)\hat{F}_{x'}(z',t')\rangle = \frac{(2\pi)^2}{AN} 2D^n_{x,x'}\,\delta(z-z')\,\delta(t-t') \tag{E2}$$

and similarly for the antinormal-ordered noise correlations

$$\langle \hat{F}_x(z,t)\hat{F}^\dagger_{x'}(z',t')\rangle = \frac{(2\pi)^2}{AN} 2D^{an}_{x,x'}\,\delta(z-z')\,\delta(t-t'). \tag{E3}$$

The normal-ordered $2D^n_{x,x'}$ and antinormal-ordered $2D^{an}_{x,x'}$ coefficients are calculated using Einstein's relation and the atomic equations for continuous variables. For the purpose of evaluating the correlations, these coefficients are expressed in terms of the steady-state matrix elements ρ^{st}_{ij} and the thermal photon number $\bar{n}_f$.

Note that when the thermal temperature is zero, the excited populations are negligible, $\rho^{st}_{dd}\simeq\rho^{st}_{aa}\simeq 0$. If the dephasings are such that $\gamma^{dep}_{dc}=\gamma^{dep}_{ad}+\gamma^{dep}_{ac}=\gamma^{dep}_{db}+\gamma^{dep}_{bc}$, the only finite diffusion coefficients are $2\tilde{D}^n_{ac,ac}=2\gamma_{ac}\rho^{st}_{cc}$, $2\tilde{D}^n_{bc,bc}=2\gamma_{bc}\rho^{st}_{cc}$, $2\tilde{D}^{an}_{bc,bc}=2\gamma_{bc}\rho^{st}_{bb}$, and $2\tilde{D}^{an}_{bd,bd}=2\gamma_{db}\rho^{st}_{bb}$. This means that the correlations due to noise operators are governed by the decoherence rates γ_{db} and γ_{ac} of the Stokes and anti-Stokes transitions as well as the decoherence γ_{bc} between the ground states.

APPENDIX F: DECORRELATION FOR GAUSSIAN NOISE

The solutions of the Stokes and anti-Stokes operators are composed of the boundary and the noise parts

$$\hat{E}_s(L,t) = \hat{B}_s(L,t) + \hat{N}_s(L,t), \tag{F1}$$

$$\hat{E}_a^\dagger(0,t) = \hat{B}_a^\dagger(L,t) + \hat{N}_a^\dagger(L,t), \tag{F2}$$

where $\hat{B}_s(L,t)$, $\hat{B}_a(L,t)$, $\hat{N}_s(L,t)$, and $\hat{N}_a^\dagger(L,t)$ are the inverse Fourier transforms of the terms in Eqs. (10) and (11).

Since the noise operators in vacuum are well known to satisfy Gaussian decorrelation and the odd products of noise operators vanish, the noise part satisfies the Gaussian decorrelation. Thus, we only need to verify one term in $G_{as}^{(2)}(\tau) = \langle \hat{E}_s^\dagger(t)\hat{E}_a^\dagger(t+\tau)\hat{E}_a(t+\tau)\hat{E}_s(t)\rangle$—i.e., the term with all boundary operators. Since the boundary parts $\hat{B}_f$ contain $\hat{E}_f(0,\nu) \propto \hat{a}_f$, we can write

$$\langle B_s^\dagger(t)B_a^\dagger(t+\tau)B_a(t+\tau)B_s(t)\rangle \propto \langle (\hat{a}_s^\dagger + \hat{a}_a)(\hat{a}_a^\dagger + \hat{a}_s) \times (\hat{a}_a + \hat{a}_s^\dagger)(\hat{a}_s + \hat{a}_a^\dagger)\rangle, \tag{F3}$$

where coefficients do not affect our argument and are left out. Straightforward expansion gives

$$2\langle \hat{n}_s\rangle^2 + 4\langle \hat{n}_s\rangle + 4\langle \hat{n}_s\rangle\langle \hat{n}_a\rangle + 2\langle \hat{n}_a\rangle^2 + 4\langle \hat{n}_a\rangle + 2 = \langle \hat{n}_s^2\rangle + 3\langle \hat{n}_s\rangle + 4\langle \hat{n}_s\rangle\langle \hat{n}_a\rangle + \langle \hat{n}_a^2\rangle + 3\langle \hat{n}_a\rangle + 2, \tag{F4}$$

where the second line follows for the thermal state, $\langle \hat{n}_s^2\rangle = 2\langle \hat{n}_s\rangle^2 + \langle \hat{n}_s\rangle$.

If we use Gaussian decorrelation on Eq. (3), we have

$$\langle (a_s^\dagger + a_a)(a_a^\dagger + a_s)\rangle\langle (a_a + a_s^\dagger)(a_s + a_a^\dagger)\rangle + \langle (a_s^\dagger + a_a)(a_a + a_s^\dagger)\rangle\langle (a_a^\dagger + a_s)(a_s + a_a^\dagger)\rangle + \langle (a_s^\dagger + a_a)(a_s + a_a^\dagger)\rangle\langle (a_a^\dagger + a_s)(a_a + a_s^\dagger)\rangle = 2(\langle \hat{n}_s\rangle + \langle \hat{n}_a\rangle + 1)^2, \tag{F5}$$

which proves that the Gaussian decorrelation applies to operators in the thermal state, as well as the vacuum state: when $\langle \hat{n}_s\rangle = \langle \hat{n}_a\rangle = 0$. Thus, the decorrelation of Eq. (27) is justified.

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$\hat{p}_{ac}(z,\nu)$, $\hat{p}_{ad}(z,\nu)$, $\hat{p}_{bc}(z,\nu)$, $\hat{p}_{bd}(z,\nu)$, $\hat{E}_s$, and $\hat{E}_a^\dagger$ simultaneously first by eliminating $\hat{p}_{bd}(z,\nu)$, $\hat{p}_{ac}(z,\nu)$, and then $\hat{p}_{ad}(z,\nu)$, $\hat{p}_{bc}(z,\nu)$, thus giving the coupled equations (1) and (2).

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