Up-conversion dynamics for temporally entangled two-photon pulses

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We analyze the up conversion of a two-photon pulse having temporal entanglement on the basis of a full quantum formalism that treats both photons and optical media quantum mechanically. We derive a formula of the up-converted photon wave function, which is applicable to arbitrary input two-photon states for a three-level system, as the simplest second-order nonlinear optical system. As the input, we employ three kinds of temporally entangled two-photon pulses: correlated, uncorrelated, and anticorrelated. We observe the up-conversion efficiency and the temporal profile of the up-converted photon. Our results reveal the crossover behavior of the up conversion from anticorrelation to correlation and show how the temporal correlation in the input is reflected in the up-conversion process.

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I. INTRODUCTION

Nonlinear optical effects appearing in single photons would be quite useful for realizing future photonic quantum technologies, including quantum information processing [1–3]. However, considerable nonlinear optical effects can be obtained only by inputting strong laser pulses containing innumerable photons. Therefore, achieving the giant optical nonlinearity sensitive to single photons is left as an open problem. On the other hand, the nonclassical nature of entangled photons may serve as a clue to overcoming the limitation of present optical technologies based on classical light. In particular, entanglements in the continuous degree of freedom (e.g., temporal, spatial, frequency, and momentum entanglements) are applicable for quantum imaging, quantum lithography, and quantum metrology [4–9]. Owing to recent experimental developments, photon sources for temporally entangled two-photon pulses [10–14] are now available, in which the temporal correlation of two photons can be engineered at will. Moreover, it has been shown theoretically that up conversion for two single-photon pulses is sensitive to multimode entanglements between single-photon pulses [15]. Therefore, it is expected that there is a possibility of a giant optical nonlinearity that is sensitive to single photons by controlling the temporal correlation of two photons.

In this paper, we reveal the uniqueness of such temporally entangled photons appearing in the optical response. As one of the simplest nonlinear optical processes, we consider how a two-photon pulse propagating in a one-dimensional space is up converted by a delta-type three-level system, which has nonvanishing transition matrix elements among three levels (see Fig. 1). Such a situation can be realized in various physical setups, such as quantum dots coupled to photonic crystal waveguides [16] and superconducting qubits coupled to a microwave transmission line [17]. As the input, three kinds of temporally entangled two-photon pulses are considered: (i) anticorrelated, (ii) uncorrelated, and (iii) correlated. In addition, we also consider a classical pulse having the mean photon number $n = 2$ as a reference. These states have the identical pulse shapes and therefore are indistinguishable by the classical measurements, which are essentially concerned with the first-order correlation of the field. On the basis of the full quantum analysis, in which both the multimode photon field and the optical media are treated quantum mechanically, we clarify how the temporal entanglement in the input pulse affects the up-conversion dynamics. We also analyze the crossover of the temporal correlation of the input in the up-conversion dynamics from anticorrelation to correlation, and specify the optimal correlation for up-conversion efficiency.

This paper is organized as follows. Details of the theoretical model are explained in Sec. II. We derive an analytical form of the propagating function in Sec. III and connect the wave functions of the up-converted photon and the input two photons. Based on this formal result, we visualize numerically the up-conversion probability and the pulse shape in Sec. IV, giving an intuitive interpretation of how the temporally entangled two photons are up converted. Section V is devoted to a summary.

II. SYSTEM

A. Nonlinear optical system

As illustrated in Fig. 1, we consider the system composed of two kinds of one-dimensional photon fields [horizontally (H) polarized and vertically (V) polarized] and a delta-type three-level system (we call hereafter an "atom") located at $r = 0$. The three atomic levels are denoted by $|g\rangle$, $|m\rangle$, and $|e\rangle$. The energies $|m\rangle$ and $|e\rangle$ measured from $|g\rangle$ are Ω_m and Ω_e, respectively, and the radiative decay rates associated with the $|e\rangle \to |g\rangle$, $|e\rangle \to |\mathrm{m}\rangle$, and $|\mathrm{m}\rangle \to |\mathrm{g}\rangle$ transitions are denoted by Γ_1, Γ_2, and Γ_3, respectively. The $|e\rangle \to |m\rangle$ and $|m\rangle \to |\mathrm{g}\rangle$ transitions are assisted by H-polarized photons, and the $|e\rangle \to |g\rangle$ transition by V-polarized photons. Employing natural units ($\hbar = c = 1$), the Hamiltonian for the overall system is given,

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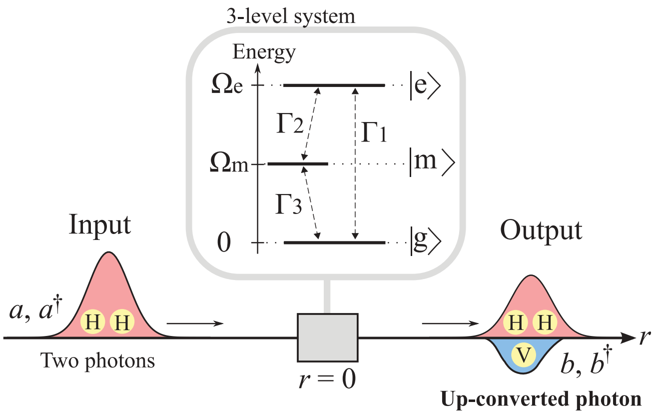

FIG. 1. (Color online) The physical setup. Two H-polarized photons are input from the input port (the $r < 0$ region). After interacting with the three-level system, an up-converted photon with V polarization is generated in the output port (the $r > 0$ region) with a certain probability.

under the rotating-wave approximation, by

$$\mathcal{H} = \Omega_e \sigma_{ee} + \Omega_m \sigma_{mm} + \int dk\, k(a_k^\dagger a_k + b_k^\dagger b_k) + \int \frac{dk}{\sqrt{2\pi}} (i\sqrt{\Gamma_1}\sigma_{eg} b_k + i\sqrt{\Gamma_2}\sigma_{em} a_k + i\sqrt{\Gamma_3}\sigma_{mg} a_k + \text{H.c.}), \tag{1}$$

where the atomic transition operators are defined as $\sigma_{eg} = |e\rangle\langle g|$, for example. The a_k (b_k) denotes the annihilation operator of the H-polarized (V-polarized) photon with the wave number k. The real-space representation $\tilde{a}_r$ of the tilde operator is defined as the Fourier transform of a_k:

$$\tilde{a}_r = \frac{1}{\sqrt{2\pi}} \int dk\, a_k e^{ikr}. \tag{2}$$

The $\tilde{b}_r$ is defined similarly.

B. Input and output photons

Initially, the atom is in the ground state and two H-polarized photons are input from the input port (the $r < 0$ region). After the photon-atom interaction, an up-converted photon with V polarization may be generated in the output port (the $r > 0$ region) with a certain probability. The state vector of the input two photons can be written as

$$|\psi_{\text{in}}\rangle = \int\int dr_1 dr_2\, \frac{f(r_1,r_2)}{\sqrt{2}} \tilde{a}_{r_1}^\dagger \tilde{a}_{r_2}^\dagger |0\rangle, \tag{3}$$

where $|0\rangle$ represents the overall ground state (the product of the atomic ground state and the photonic vacuum state), and $f(r_1,r_2)$ is the two-photon wave function, which is normalized as $\int dr_1 dr_2 |f(r_1,r_2)|^2 = 1$ and symmetrized as $f(r_1,r_2) = f(r_2,r_1)$. Note that the input two photons are initially localized at an initial position, which means $f(r_1,r_2) = 0$ for $r_1 > 0$ or $r_2 > 0$. The state vector of the output photons is determined by the Schrödinger equation

$$|\psi_{\text{out}}\rangle = \exp(-i\mathcal{H}t)|\psi_{\text{in}}\rangle, \tag{4}$$

where the final moment t is a sufficiently large time at which the atom is completely de-excited. The output state may contain a V-polarized single-photon component associated with the $|e\rangle \to |g\rangle$ transition. The output state vector can thus be written as

$$|\psi_{\text{out}}\rangle = \int\int dr_1 dr_2 \frac{g(r_1,r_2;t)}{\sqrt{2}} \tilde{a}_{r_1}^\dagger \tilde{a}_{r_2}^\dagger |0\rangle + \int dr\, h(r;t) \tilde{b}_r^\dagger |0\rangle, \tag{5}$$

where $g(r_1,r_2;t)$ and $h(r;t)$ are the H-polarized two-photon and V-polarized single-photon wave functions, respectively. Note that $g(r_1,r_2;t)$ and $h(r;t)$ vanish on the input port. The probabilities for up conversion and for the absence of up conversion are given as the norm of $h(r;t)$ and $g(r_1,r_2;t)$, respectively, by

$$P_1 = \int dr |h(r;t)|^2, \tag{6}$$

$$P_2 = \int\int dr_1 dr_2 |g(r_1,r_2;t)|^2. \tag{7}$$

These probabilities satisfy $P_1 + P_2 = 1$, and are independent of the final moment t as long as t is sufficiently large.

III. RELATION BETWEEN INPUT AND UP-CONVERTED PHOTONS

In the following part of this paper, we analyze the up-conversion dynamics of temporally entangled two-photon pulses. In this section, we derive the analytical expression for the wave function $h(r;t)$ of the up-converted photon from the Schrödinger equation (4) [18,19].

A. Propagator for up-conversion dynamics

The wave function of the up-converted photon can be written as

$$h(r;t) = \langle 0|\tilde{b}_r|\psi_{\text{out}}\rangle = \langle 0|\tilde{b}_r(t)|\psi_{\text{in}}\rangle = \int\int dr_1' dr_2' G(r,r_1',r_2';t) f(r_1',r_2'), \tag{8}$$

where $A(t) = e^{i\mathcal{H}t} A e^{-i\mathcal{H}t}$ (Heisenberg picture), and $G(r,r_1',r_2';t)$ is the propagator related to the up-converted photon, which is defined, for $r' < 0 < r$, by

$$G(r,r_1',r_2';t) = \frac{1}{\sqrt{2}} \langle 0|\tilde{b}_r(t) \tilde{a}_{r_1'}^\dagger \tilde{a}_{r_2'}^\dagger |0\rangle. \tag{9}$$

This propagator contains complete information about the up-conversion dynamics of the input two photons. Note that Eq. (8) holds for an arbitrary input two-photon wave function $f(r_1,r_2)$.

B. Up-converted photon wave function

As shown in the Appendix, the propagator (9) is given, for $0 < r < t$, by

$$G(r,r_1',r_2';t) = -\sqrt{\frac{\Gamma_1\Gamma_2\Gamma_3}{2}} \int_0^{t-r} d\tau_1 \int_0^{\tau_1} d\tau_2\, [\delta(\tau_1 + r_2')\delta(\tau_2 + r_1') + \delta(\tau_1 + r_1')\delta(\tau_2 + r_2')]$$

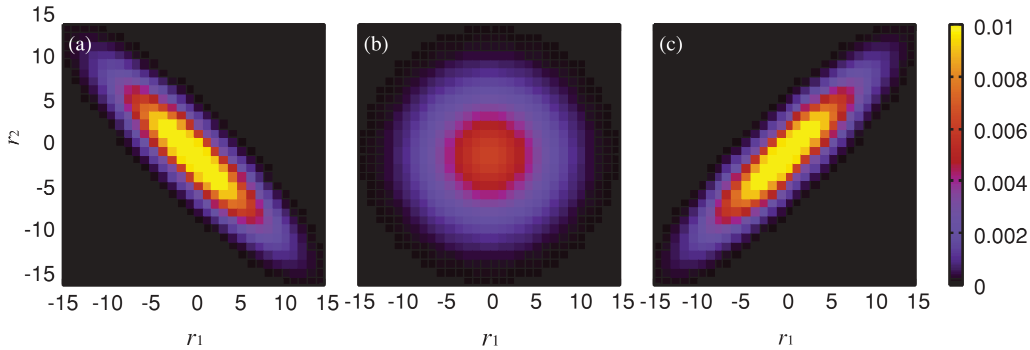

FIG. 2. (Color online) Density plots of $R^{(2)}(r_1,r_2)$ for the input two-photon wave function in this paper: (a) temporally anticorrelated ($\rho = -0.9$), (b) temporally uncorrelated ($\rho = 0$), and (c) temporally correlated ($\rho = 0.9$) states.

$$\times \exp\left\{-\left[\left(i\Omega_e + \frac{\Gamma_1+\Gamma_2}{2}\right)(t-r-\tau_1)\right]\right\}$$
$$\times \exp\left[-\left(i\Omega_m + \frac{\Gamma_3}{2}\right)(\tau_1-\tau_2)\right]. \quad (10)$$

From Eqs. (8) and (10), the wave function $h(r;t)$ of the up-converted photon is given, in terms of the input wave function $f(r_1,r_2)$, by

$$h(r;t) = -\sqrt{2\Gamma_1\Gamma_2\Gamma_3}\exp\left[-\left(i\Omega_e + \frac{\Gamma_1+\Gamma_2}{2}\right)(t-r)\right]$$
$$\times \int_0^{t-r} d\tau_1 \int_0^{\tau_1} d\tau_2\ f(-\tau_1,-\tau_2)$$
$$\times \exp\left\{\left[i(\Omega_e-\Omega_m) + \frac{\Gamma_1+\Gamma_2-\Gamma_3}{2}\right]\tau_1\right.$$
$$\left. + \left(i\Omega_m + \frac{\Gamma_3}{2}\right)\tau_2\right\}. \quad (11)$$

This formula is applicable to arbitrary input two-photon states. In fact, it is the same expression as the up-converted photon wave functions that are derived for some specific inputs in Ref. [15].

IV. RESULTS

A. Input two-photon wave function

As the input two-photon wave function, we employ the following bivariate Gaussian:

$$f(r_1,r_2) = \left[\frac{2}{\pi(1-\rho^2)^{1/2}\,l^2}\right]^{1/2}$$
$$\times \exp\left[-\frac{\bar{r}_1^2+\bar{r}_2^2-2\rho\bar{r}_1\bar{r}_2}{(1-\rho^2)\,l^2} + i\Omega(\bar{r}_1+\bar{r}_2)\right], \quad (12)$$

where $\bar{r} = r - a$, and a represents the initial position, which is a redundant parameter provided that $|a| \gg l$, as is assumed here. The parameters ρ, l, and Ω denote the correlation between the two photons, the pulse length, and the photon frequency, respectively. The correlation parameter ρ lies in the $-1 < \rho < 1$ region, where positive (negative) values indicate the temporal correlation (anticorrelation) of two photons, and $\rho = 0$ implies no correlation. We can analyze the crossover behavior of the up conversion from anticorrelation to correlation by continuously changing the value of ρ. Figure 2 shows the density plots of the second-order correlation function $R^{(2)}(r_1,r_2)$ defined by

$$R^{(2)}(r_1,r_2) = \frac{\langle\psi_{\rm in}|\tilde{a}_{r_2}^\dagger\tilde{a}_{r_1}^\dagger\tilde{a}_{r_1}\tilde{a}_{r_2}|\psi_{\rm in}\rangle}{2} = |f(r_1,r_2)|^2. \quad (13)$$

It is observed that two photons tend to be found at the same position as ρ is increased. Note that the single-photon intensity $I(r) \equiv \langle\psi_{\rm in}|\tilde{a}_r^\dagger\tilde{a}_r|\psi_{\rm in}\rangle/2 = \int dr'|f(r,r')|^2 = (2/\pi l^2)^{1/2}\exp(-2\bar{r}^2/l^2)$ is independent of ρ. Therefore, two-photon states for different values of ρ are indistinguishable by measurements of the first-order correlations of the photon field.

For reference, we also consider a classical light pulse with the mean photon number $n = 2$:

$$|\psi_{\rm in}^C\rangle = \mathcal{N}\exp\left[\sqrt{2}\int dr f_C(r)\tilde{a}_r^\dagger\right]|0\rangle, \quad (14)$$

where $f_C(r) = \sqrt{I(r)}\exp(i\Omega\bar{r})$, and $\mathcal{N} = e^{-1}$ is the normalization factor. Note that $|\psi_{\rm in}^C\rangle$ has the same intensity as the single-photon intensity $I(r)$ for $f(r_1,r_2)$. For this input state, the up-conversion probability is given by

$$P_1^C = \int dr\langle\psi_{\rm in}^C|\tilde{b}_r^\dagger(t)\tilde{b}_r(t)|\psi_{\rm in}^C\rangle, \quad (15)$$

which can be calculated from the input-output relation of $\tilde{b}_r$ and the equations of motion for the atomic coherence.

In the following subsections, we investigate the up-conversion dynamics for the input two-photon state (12) by numerically calculating the analytical expression (11) for the up-converted photon. For simplicity, we employ the following assumptions: (i) Regarding the atomic energy, $\Omega_m = \Omega_e/2$ ($= \Omega_0$); (ii) regarding the atomic decay rates, $\Gamma_1 = \Gamma_2 = \Gamma_3$ ($= \Gamma$), and Γ^{-1} is used as the unit of time and length; and (iii) the two input photons are resonant to the $|g\rangle \to |m\rangle$ and $|m\rangle \to |e\rangle$ transitions $\Omega_0 = \Omega$.

B. Up-conversion probability

First, we investigate the up-conversion probability P_1, which is given by Eq. (6) as the norm of $h(r;t)$. Figure 3 shows the crossover behavior of the up conversion from anticorrelation to correlation, where the up-conversion probability P_1 is plotted as a function of the correlation parameter ρ (solid

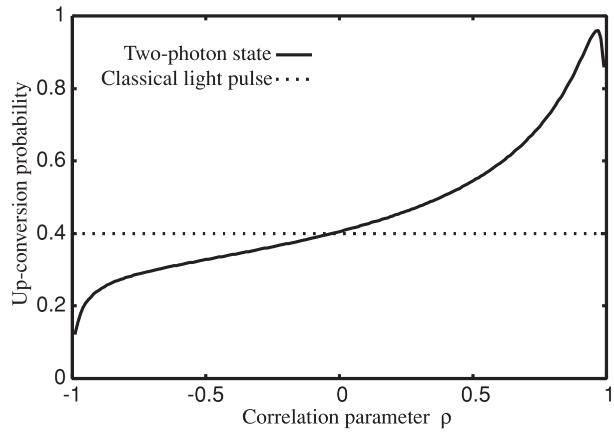

FIG. 3. Crossover behavior of the up conversion. The solid line shows the up-conversion probability as a function of ρ, where the pulse length is fixed at $l = 10\Gamma^{-1}$. The dotted line shows the probability for the classical light pulse with the mean photon number $n = 2$. The up-conversion probability is maximized ($P_1 \simeq 0.96$) at $\rho \simeq 0.97$.

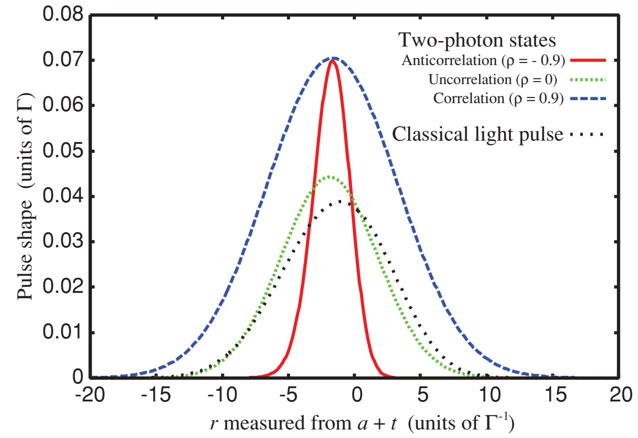

FIG. 4. (Color online) Pulse shapes of the up-converted photon. The pulse length is fixed at $l = 10\Gamma^{-1}$. Results for the anticorrelated ($\rho = -0.9$), uncorrelated ($\rho = 0$), and correlated ($\rho = 0.9$) two-photon states are indicated by the solid, fine dotted, and dashed lines, respectively. The dotted line shows the output pulse shape for the classical input.

line) by fixing the pulse length l at $10\Gamma^{-1}$. As a reference, the probability for the classical light pulse, which is given by Eq. (15), is also plotted (dotted line in Fig. 3). It is observed that the up-conversion probability tends to increase as ρ is increased, and is maximized ($P_1 \simeq 0.96$) at $\rho \simeq 0.97$. The up-conversion probabilities for the temporally uncorrelated state ($\rho = 0$) and for the reference classical pulse almost coincide, $P_1 \simeq P_1^C \simeq 0.4$. For temporally correlated input states, we can efficiently obtain the up-converted photon. The up-converted photon can be basically obtained with higher efficiency for larger correlation ρ, because the first excitation ($|g\rangle \to |m\rangle$) should be followed immediately by the second excitation ($|m\rangle \to |e\rangle$) before the relaxation ($|m\rangle \to |g\rangle$). However, the efficiency is decreased in the $\rho \to 1$ limit, in which two photons arrive at the atom exactly at the same moment; this is because the atom does not absorb two photons within a much shorter time than the photon-atom interaction time Γ^{-1}. This behavior is quite similar to that of the optical nonlinearity calculated in Ref. [19], where a H-polarized and a V-polarized photon interact at a V-type three-level system.

C. Pulse shape of up-converted photon

Next, we examine the the pulse shape of the up-converted photon, which is given by

$$I_{\mathrm{out}}(r;t) = \langle \psi_{\mathrm{in}} | \tilde{b}_r^\dagger(t) \tilde{b}_r(t) | \psi_{\mathrm{in}} \rangle = |h(r;t)|^2. \tag{16}$$

For the temporally anticorrelated ($\rho = -0.9$), uncorrelated ($\rho = 0$), and correlated ($\rho = 0.9$) two-photon states, the pulse shapes of the up-converted photon are plotted in Fig. 4. It is observed that the length of the up-converted photon pulse for the anticorrelated input (solid line in Fig. 4) is shorter than that for the other input states. In contrast, we obtain a long pulse for the correlated input state (dashed line in Fig. 4), which indicates that the correlated two photons induce a strong nonlinear effect. The pulse shape for the uncorrelated input state (fine dotted line in Fig. 4) is similar to that for the classical light pulse (dotted line in Fig. 4). The slight difference between them is due to the fact that the classical light pulse has not only the two-photon component, but also the one-photon and the more-than-two-photons components.

D. Intuitive explanation

The up-conversion dynamics of temporally entangled two-photon pulses can intuitively be understood by the following interpretation: the up conversion arises in the diagonal area $|r_1 - r_2| \lesssim \Gamma^{-1}$ of the second-order correlation $R^{(2)}(r_1,r_2)$, where two photons in the pulse are absorbed by the atom within the time interval Γ^{-1}. The pulse length of the up-converted photon is determined by the width of $R^{(2)}$ on the diagonal line $r_1 = r_2$. The temporally correlated state for which R_2 is large near the diagonal line [Fig. 2(c)] gives a large up-conversion probability [Fig. 3]. The resultant up-converted photon has a long pulse length [Fig. 4]. In contrast, the temporally anticorrelated state for which R_2 is large near the antidiagonal line [Fig. 2(a)] gives a small up-conversion probability. The pulse length of the resultant up-converted photon becomes shorter than that for the uncorrelated state.

V. SUMMARY

We have investigated theoretically the up-conversion dynamics of temporally entangled two-photon pulses, assuming a three-level atom as the simplest second-order nonlinear optical system. We have derived the formula of the up-converted photon wave function [Eq. (11)], which is applicable to arbitrary input two-photon states. Temporal entanglements of the input two-photon pulses are characterized by the correlation parameter ρ, as shown in Fig. 2. From the up-converted photon wave function (11), we have numerically evaluated the up-conversion probability, where the crossover behavior from anticorrelation to correlation is clarified by continuously changing the value of ρ and the optimal correlation is specified, as shown in Fig. 3. We have also visualized the pulse shape of the up-converted photon in Fig. 4. It is found that, although the input states are indistinguishable by the first-order correlation, the up-conversion efficiency depends drastically on the temporal correlation ρ. The dependence of the up-conversion dynamics on ρ can intuitively be explained by considering that the up conversion arises in the diagonal area $|r_1 - r_2| \lesssim \Gamma^{-1}$ of the second-order correlation $R^{(2)}(r_1,r_2)$. The obtained results indicate that a temporally correlated (anticorrelated) two-photon pulse has an effectively longer (shorter) pulse length than that of the uncorrelated two-photon pulse on the up-conversion dynamics. We believe that controlling the temporal entanglements of a two-photon pulse opens up new possibilities of photonic quantum technologies.

ACKNOWLEDGMENT

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APPENDIX: DERIVATION OF THE PROPAGATOR

In this appendix, we derive the propagator (9). For future convenience, we introduce a coherent state given by

$$|\phi\rangle = \mathcal{N}_\phi \exp\left(\sum_{j=1,2} \mu_j \tilde{a}^\dagger_{r'_j}\right)|0\rangle, \tag{A1}$$

where μ_j are perturbation coefficients and $\mathcal{N}_\phi$ is a normalization factor. Note that $\tilde{a}_r|\phi\rangle = \sum_{j=1,2}\mu_j\delta(r-r'_j)|\phi\rangle$ and $\tilde{b}_r|\phi\rangle = 0$. From the Heisenberg equations for the photonic operators, the output fields are obtained, for $0 \leqslant r < \tau \leqslant t$, as

$$\tilde{a}_r(\tau) = \tilde{a}_{r-\tau}(0) - [\sqrt{\Gamma_2}\sigma_{me}(\tau - r) + \sqrt{\Gamma_3}\sigma_{gm}(\tau - r)]\,\theta(r), \tag{A2}$$

$$\tilde{b}_r(\tau) = \tilde{b}_{r-\tau}(0) - \sqrt{\Gamma_1}\sigma_{ge}(\tau - r)\,\theta(r), \tag{A3}$$

where $\theta(r)$ is the Heaviside step function. From Eqs. (A1) and (A3), the propagator can be written as

$$\begin{aligned} G(r,r'_1,r'_2;t) &= \frac{1}{\sqrt{2}}\langle\tilde{b}_r(t)\rangle^{(\mu_1\mu_2)} \\ &= -\sqrt{\frac{\Gamma_1}{2}}\langle\sigma_{ge}(t-r)\rangle^{(\mu_1\mu_2)}\theta(r), \end{aligned} \tag{A4}$$

where $\langle\tilde{b}_r(t)\rangle^{(\mu_1\mu_2)}$ is the second-order component of $\langle\tilde{b}_r(t)\rangle = \langle\phi|\tilde{b}_r(t)|\phi\rangle$ proportional to $\mu_1\mu_2$, for example.

From the Heisenberg equations for σ_{ge} and σ_{gm}, the equations of motion for $\langle\sigma_{ge}\rangle$ and $\langle\sigma_{gm}\rangle$ are given by

$$\begin{aligned} \frac{d}{d\tau}\langle\sigma_{ge}(\tau)\rangle &= -\left(i\Omega_e + \frac{\Gamma_1+\Gamma_2}{2}\right)\langle\sigma_{ge}(\tau)\rangle + \sum_{j=1,2}\mu_j\delta(\tau + r_j) \\ &\quad\times[\sqrt{\Gamma_2}\langle\sigma_{gm}(\tau)\rangle - \sqrt{\Gamma_3}\langle\sigma_{me}(\tau)\rangle], \end{aligned} \tag{A5}$$

$$\begin{aligned} \frac{d}{d\tau}\langle\sigma_{gm}(\tau)\rangle &= -\left(i\Omega_m + \frac{\Gamma_3}{2}\right)\langle\sigma_{gm}(\tau)\rangle + \sqrt{\Gamma_2\Gamma_3}\langle\sigma_{me}(\tau)\rangle \\ &\quad - \sum_{j=1,2}\mu_j^*\delta(\tau + r_j)\sqrt{\Gamma_2}\langle\sigma_{ge}(\tau)\rangle \\ &\quad - \sum_{j=1,2}\mu_j\delta(\tau + r_j)\sqrt{\Gamma_3}[\langle\sigma_{mm}(\tau)\rangle - \langle\sigma_{gg}(\tau)\rangle], \end{aligned} \tag{A6}$$

where we used the relations of Eqs. (A2) and (A3), and the virtue of the coherent state $|\phi\rangle$: $\tilde{a}_r|\phi\rangle = \sum_{j=1,2}\mu_j\delta(r - r'_j)|\phi\rangle$ and $\tilde{b}_r|\phi\rangle = 0$. Using the rotating-wave approximation and expanding these equations in powers of μ_1 and μ_2, it is found that, except for $\langle\sigma_{ge}\rangle^{(\mu_1\mu_2)}$ and $\langle\sigma_{gm}\rangle^{(\mu_j)}$, the components of the expectations of the atomic operators have no contribution to the equation of motion for $\langle\sigma_{ge}\rangle^{(\mu_1\mu_2)}$. The equations of motion for $\langle\sigma_{ge}\rangle^{(\mu_1\mu_2)}$ and $\langle\sigma_{gm}\rangle^{(\mu_j)}$ can be written as follows:

$$\begin{aligned} \frac{d}{d\tau}\langle\sigma_{ge}(\tau)\rangle^{(\mu_1\mu_2)} &= -\left(i\Omega_e + \frac{\Gamma_1+\Gamma_2}{2}\right)\langle\sigma_{ge}(\tau)\rangle^{(\mu_1\mu_2)} \\ &\quad + \sqrt{\Gamma_2}[\delta(\tau + r_2)\langle\sigma_{gm}(\tau)\rangle^{(\mu_1)} + \delta(\tau + r_1)\langle\sigma_{gm}(\tau)\rangle^{(\mu_2)}], \end{aligned} \tag{A7}$$

$$\frac{d}{d\tau}\langle\sigma_{gm}(\tau)\rangle^{(\mu_j)} = -\left(i\Omega_m + \frac{\Gamma_3}{2}\right)\langle\sigma_{gm}(\tau)\rangle^{(\mu_j)} + \sqrt{\Gamma_3}\delta(\tau + r_j). \tag{A8}$$

Solving these equations with the initial conditions $\langle\sigma_{ge}(0)\rangle^{(\mu_1\mu_2)} = 0$ and $\langle\sigma_{gm}(0)\rangle^{(\mu_j)} = 0$, the propagator (A4) is reduced, for $0 < r < t$, to Eq. (10).

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