Theoretical method for analyzing quantum dynamics of correlated photons

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We present a theoretical method for the efficient analysis of quantum nonlinear dynamics of correlated photons. Since correlated photons can be regarded as a superposition of uncorrelated photons, semiclassical analysis can be applied to this problem. The proposed method is demonstrated for a V-type three-level atom as a nonlinear optical system.

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I. INTRODUCTION

Control of a photon by another photon using nonlinear optical effects has broad applicability in quantum information processing [1–5]. Therefore, quantitative nonlinear-optics theory applicable to the single-photon domain is of great importance. The nonlinear dynamics of photons has conventionally been discussed under the single-mode approximation, in which the photonic mode functions are assumed to be unchanged during the nonlinear processes [6]. However, this assumption is valid only when the input photons are off-resonant and therefore the nonlinear effects are weak. In order to develop a quantitative theory applicable to resonant input photons, we must treat both photons and optical media as active quantum-mechanical degrees of freedom (full-quantum formalism) while adopting a multimode nature for the photon field. The problem with this rigorous treatment as a quantum many-particle problem is the intensive numerical computation involved [7,8].

In order to reduce the computational burden in multimode quantum-optics theory, we have developed a semiclassical method for solving the dynamics of photons [9,10]. Since this method relies strongly on the relationship between photons and classical light pulses, the applicability of the method is limited to temporally or spatially *uncorrelated* input photons, which constitutes classical light pulses. However, this method is not applicable to the problem of *correlated* input photons, which are generated by nonclassical light sources. For example, the signal and idler photons generated by spontaneous parametric down-conversion are correlated strongly in time, since they are generated almost simultaneously from a parent photon [11–13]. The objective of the present Brief Report is to present a method for analyzing the dynamics of such correlated photons. The key idea is that correlated photons can be regarded as a superposition of uncorrelated input photons. The rest of this Brief Report is organized as follows. The basic idea and general formula are presented in Sec. II. In Sec. III, these formulas are applied to a concrete problem in which two correlated photons interact at a V-type three-level atom. Finally, in Sec. IV, extension of the proposed method is briefly discussed.

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II. METHOD

A. State vector of input and output photons

In this study, we investigate the nonlinear interaction of spatially correlated photons. As illustrated in Fig. 1, m horizontally (H) polarized photons and n vertically (V) polarized photons are inputted from the input port ($r<0$ region), interact with a nonlinear medium (at $r\simeq 0$), and are outputted into the output port ($r>0$ region). For simplicity in presentation, we assume that the number of photons is conserved during the interaction. (However, extending the treatment to non-conserving processes such as up-conversion is straightforward; see Sec. IV.) The state vector of the input photons is given by

$$|\psi_{\rm in}\rangle = \int_{-\infty}^{0} d^{m+n}\mathbf{r}\frac{f(\mathbf{r})}{\sqrt{m\,!\,n!}}a_{r_1}^\dagger\cdots a_{r_m}^\dagger b_{r_{m+1}}^\dagger\cdots b_{r_{m+n}}^\dagger|0\rangle, \quad (1)$$

where $a_r^\dagger$ and $b_r^\dagger$ are the photon creation operators in the real-space representation for the H and V polarized fields, the set of $m+n$ space variables $(r_1,\ldots,r_{m+n})$ is abbreviated as $\mathbf{r}$, and $f(\mathbf{r})$ is the spatial wave function of the input photons, normalized as $\int_{-\infty}^{0}d^{m+n}\mathbf{r}|f|^2=1$. The output state is determined by the Schrödinger equation,

$$|\psi_{\rm out}\rangle = e^{-i\mathcal{H}t}|\psi_{\rm in}\rangle. \quad (2)$$

Under the assumption of photon-number conservation, the output state vector can be written as

$$|\psi_{\rm out}\rangle = \int_{0}^{\infty} d^{m+n}\mathbf{r}\frac{g(\mathbf{r};t)}{\sqrt{m\,!\,n!}}a_{r_1}^\dagger\cdots a_{r_m}^\dagger b_{r_{m+1}}^\dagger\cdots b_{r_{m+n}}^\dagger|0\rangle, \quad (3)$$

where $g(\mathbf{r};t)$ is the output wave function, normalized as $\int_0^\infty d^{m+n}\mathbf{r}|g|^2=1$.

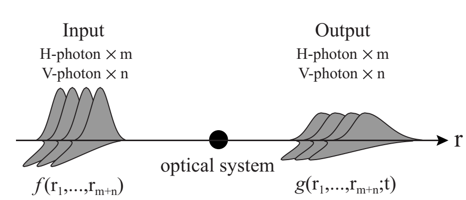

FIG. 1. Physical situation discussed in Sec. II. m H polarized photons and n V polarized photons are input into a nonlinear optical system. The number of photons is assumed to be conserved between the input and output.

In earlier studies, we assumed that the input photons are *uncorrelated* [9,10]. Namely, the input wave function is given by a product of single-photon wave functions, as $f(\mathbf{r})=\Pi_{j=1}^{m+n}\xi_j(r_j)$. For such cases, by considering *classical* input pulses, the output state can be calculated effectively with the aid of semiclassical analysis, bypassing full-quantum analysis. This is because the target input state $|\psi_{\rm in}\rangle$ is contained in the classical light pulse as the $(m+n)$th-order component [9,10]. In contrast, in this study, we treat the case of *correlated* input photons, in which the input wave function cannot be factored. At a glance, the semiclassical analysis is no longer applicable to this problem, since such correlated photons have no classical counterpart. However, although the input state $|\psi_{\rm in}\rangle$ itself has no classical counterpart, $a_{r_1}^\dagger\cdots b_{r_{m+n}}^\dagger|0\rangle$ in $|\psi_{\rm in}\rangle$ is an uncorrelated state and has a classical counterpart. This fact implies that the semiclassical analysis is applicable in deriving the propagator of photons, defined in Sec. II B.

B. Definition of multiple-photon propagator

The $(m+n)$-photon propagator is defined as the wave function of $m+n$ photons, which are initially located at $\mathbf{r}'=(r_1',\ldots,r_{m+n}')$. The $(m+n)$-photon propagator is defined by

$$G_{m+n}(\mathbf{r},\mathbf{r}';t)=\frac{\langle 0|a_{r_1}(t)\cdots b_{r_{m+n}}(t)a_{r_1'}^\dagger\cdots b_{r_{m+n}'}^\dagger|0\rangle}{m\,!\,n!}, \quad (4)$$

where $A(t)=e^{i\mathcal{H}t}Ae^{-i\mathcal{H}t}$ (Heisenberg picture). From Eqs. (1)–(3), we obtain the following equation connecting the input and output wave functions:

$$g(\mathbf{r};t)=\int_{-\infty}^{0}d^{m+n}\mathbf{r}'G_{m+n}(\mathbf{r},\mathbf{r}';t)f(\mathbf{r}'). \quad (5)$$

It is of note that Eq. (5) holds for general input photons, including correlated ones. The propagator G_{m+n} contains complete information on the nonlinear dynamics of the $m+n$ input photons.

C. Calculation of the propagator by classical field

The propagator G_{m+n} describes the temporal evolution of the $(m+n)$-photon state, $a_{r_1'}^\dagger\cdots b_{r_{m+n}'}^\dagger|0\rangle$. However, instead of considering this state directly, it is more convenient to consider a classical field, the state vector of which is given by

$$|\phi\rangle=\mathcal{N}\exp\left(\sum_{j=1}^{m}\mu_j a_{r_j'}^\dagger+\sum_{j=m+1}^{m+n}\mu_j b_{r_j'}^\dagger\right)|0\rangle, \quad (6)$$

where μ_j are perturbation coefficients and $\mathcal{N}=\exp(-\Sigma_j|\mu_j|^2/2)$ is a normalization constant. This state is an eigenstate of the initial field operators a_r and b_r, satisfying $a_r|\phi\rangle=\mathcal{F}_H(r)|\phi\rangle$ and $b_r|\phi\rangle=\mathcal{F}_V(r)|\phi\rangle$, where $\mathcal{F}_H(r)$ $[=\Sigma_{j=1}^{m}\mu_j\delta(r-r_j')]$ and $\mathcal{F}_V(r)$ $[=\Sigma_{j=m+1}^{m+n}\mu_j\delta(r-r_j')]$ respectively represent the field amplitudes in the H and V polarizations.

In the following part of this study, we analyze the optical response to this classical field perturbatively, namely, in powers of μ_j. In particular, we investigate the $(m+n)$-point function, $\langle\phi|a_{r_1}(t)\cdots b_{r_{m+n}}(t)|\phi\rangle$. Many components appear by expanding $|\phi\rangle$ ($\langle\phi|$) in powers of μ_j (μ_j^*). However, we focus only on the component having the perturbation coefficients of $\mu_1\cdots\mu_{m+n}$. When considering this component, $\langle\phi|$ can be replaced with $\langle 0|$, since other components in $\langle\phi|$ necessarily contain conjugated quantities, μ_j^*. On the other hand, $|\phi\rangle$ can be replaced with $a_{r_1'}^\dagger\cdots b_{r_{m+n}'}^\dagger|0\rangle$, as confirmed by expanding Eq. (6). From these observations and Eq. (4), the propagator G_{m+n} is given by

$$G_{m+n}(\mathbf{r},\mathbf{r}';t)=\frac{\langle a_{r_1}(t)\cdots b_{r_{m+n}}(t)\rangle^{\mu_1\cdots\mu_{m+n}}}{m\,!\,n!}, \quad (7)$$

where $\langle a_{r_1}(t)\cdots b_{r_{m+n}}(t)\rangle^{\mu_1\cdots\mu_{m+n}}$ denotes the component of $\langle\phi|a_{r_1}(t)\cdots b_{r_{m+n}}(t)|\phi\rangle$ having the perturbation coefficients of $\mu_1\cdots\mu_{m+n}$. As will be demonstrated in Sec. III, the classical nature of $|\phi\rangle$ considerably simplifies calculation of G_{m+n}. Equations (5)–(7) constitute the main result of the current study.

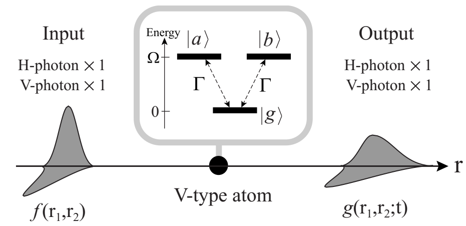

FIG. 2. Physical situation analyzed in Sec. III. A H polarized photon and a V polarized photon interact at a V-type three-level atom.

III. EXAMPLE

A. System

In this section, we demonstrate the method outlined in Sec. II by considering a specific physical situation. We investigate the interaction of a H polarized photon and a V polarized photon (the $m=n=1$ case of Sec. II) mediated by a V-type three-level atom located at $r=0$. The level structure of the atom is shown in Fig. 2. The $|g\rangle\leftrightarrow|a\rangle$ and $|g\rangle\leftrightarrow|b\rangle$ transitions are assisted by H polarized and V polarized photons, respectively. The Hamiltonian for the atom-photon system is given, taking $\hbar=c=1$, by

$$\mathcal{H}=\Omega(\sigma_{aa}+\sigma_{bb})+\int dk(k\tilde{a}_k^\dagger\tilde{a}_k+k\tilde{b}_k^\dagger\tilde{b}_k)$$
$$+i\sqrt{\Gamma}(\sigma_{ag}a_{r=0}+\sigma_{bg}b_{r=0}-\text{H.c.}), \quad (8)$$

where $\tilde{a}_k$ is the field operator in the wave-number representation $[\tilde{a}_k=(2\pi)^{-1/2}\int dr a_r e^{-ikr}]$, $\sigma_{mn}=|m\rangle\langle n|$ $(m,n=g,a,b)$, and Ω and Γ are the resonant energy and the natural linewidth of the excited states, $|a\rangle$ and $|b\rangle$.

The Heisenberg equations for the atomic operators are given by

$$\frac{d}{dt}\sigma_{ga} = -i\tilde{\Omega}\sigma_{ga} + \sqrt{\Gamma}(1-2\sigma_{aa}-\sigma_{bb})a_{-t}(0) - \sqrt{\Gamma}\sigma_{ba}b_{-t}(0), \tag{9}$$

$$\frac{d}{dt}\sigma_{gb} = -i\tilde{\Omega}\sigma_{gb} - \sqrt{\Gamma}\sigma_{ab}a_{-t}(0) + \sqrt{\Gamma}(1-\sigma_{aa}-2\sigma_{bb})b_{-t}(0), \tag{10}$$

where $\tilde{\Omega}=\Omega-i\Gamma/2$. The output field is given, for $0<r<t$, by

$$a_r(t) = a_{r-t}(0) - \sqrt{\Gamma}\sigma_{ga}(t-r), \tag{11}$$

$$b_r(t) = b_{r-t}(0) - \sqrt{\Gamma}\sigma_{gb}(t-r). \tag{12}$$

Note that Eqs. (9)–(12) can be derived from the Hamiltonian in Eq. (8).

B. Calculation of propagator

From Eqs. (6) and (7), the two-photon propagator corresponding to the current problem is given by

$$G_2(\mathbf{r},\mathbf{r}';t) = \langle a_{r_1}(t)b_{r_2}(t)\rangle^{\mu\nu}, \tag{13}$$

where $\mathbf{r}=(r_1,r_2)$ and $\mathbf{r}'=(r_1',r_2')$, and the average is taken over the following classical state:

$$|\phi\rangle = \mathcal{N}\exp(\mu a_{r_1'}^\dagger + \nu b_{r_2'}^\dagger)|0\rangle. \tag{14}$$

Here, we note the following two properties: (i) $a_r(0)|\phi\rangle = \mu\delta(r-r_1')|\phi\rangle$ and $b_r(0)|\phi\rangle=\nu\delta(r-r_2')|\phi\rangle$, and (ii) $\sigma_{mn}(t)$ commutes with $a_r(0)$ and $b_r(0)$, if $r<-t$ [14]. Substituting Eqs. (11) and (12) into Eq. (13) and using the above properties, G_2 is cast as the sum of linear and nonlinear terms as

$$G_2(\mathbf{r},\mathbf{r}';t) = G_H(r_1,r_1';t)G_V(r_2,r_2';t) + G_{NL}(\mathbf{r},\mathbf{r}';t), \tag{15}$$

where

$$G_H(r,r';t) = \delta(r-t-r') - \sqrt{\Gamma}\langle\sigma_{ga}(t-r)\rangle^{\mu}, \tag{16}$$

$$G_V(r,r';t) = \delta(r-t-r') - \sqrt{\Gamma}\langle\sigma_{gb}(t-r)\rangle^{\nu}, \tag{17}$$

$$G_{NL}(\mathbf{r},\mathbf{r}';t) = \Gamma T\langle\sigma_{ga}(t-r_1),\sigma_{gb}(t-r_2)\rangle^{\mu\nu} \tag{18}$$

where $\langle A,B\rangle=\langle AB\rangle-\langle A\rangle\langle B\rangle$ and T in the last term means time ordering.

First, we investigate the one-photon propagator, G_H. The equation of motion for $\langle\sigma_{ga}\rangle$ is obtained, from Eqs. (9) and (14), by

$$\frac{d}{dt}\langle\sigma_{ga}\rangle = -i\tilde{\Omega}\langle\sigma_{ga}\rangle + \mu\sqrt{\Gamma}\delta(t+r_1')(1-2\langle\sigma_{aa}\rangle-\langle\sigma_{bb}\rangle) - \nu\sqrt{\Gamma}\delta(t+r_2')\langle\sigma_{ba}\rangle. \tag{19}$$

We are concerned only with the quantities of the order $O(\mu^1)$ [see Eq. (16)]. However, since $\langle\sigma_{aa}\rangle\sim O(|\mu|^2)$, $\langle\sigma_{bb}\rangle\sim O(|\nu|^2)$, and $\langle\sigma_{ba}\rangle\sim O(\nu^*\mu)$, terms containing $\langle\sigma_{aa}\rangle$, $\langle\sigma_{bb}\rangle$, and $\langle\sigma_{ba}\rangle$ give higher-order contributions and are therefore irrelevant. Therefore, $\langle\sigma_{ga}(t)\rangle^{\mu}$ is determined by

$$\frac{d}{dt}\langle\sigma_{ga}(t)\rangle^{\mu} = -i\tilde{\Omega}\langle\sigma_{ga}(t)\rangle^{\mu} + \sqrt{\Gamma}\delta(t+r_1'), \tag{20}$$

with the initial condition $\langle\sigma_{ga}(0)\rangle^{\mu}=0$. Note that the field operator $a_r(0)$ can be replaced with a c-number $\delta(r-r_1')$ due to the classical property of $|\phi\rangle$, and Eq. (20) is reduced to a semiclassical equation. From these equations, we have

$$G_H(r,r';t) = \delta(r-r'-t) - \Gamma e^{i\tilde{\Omega}(r-r'-t)}\theta(r'+t-r). \tag{21}$$

The first term represents the noninteracting component, whereas the second term represents the emission from the atom. It is readily confirmed that $G_V(r,r';t)=G_H(r,r';t)$.

Next, we investigate the nonlinear part, G_{NL}. The equation of motion of $\langle\sigma_{ga}(t)\sigma_{gb}(t')\rangle^{\mu\nu}$ for $t>t'$ is obtained, again, from Eqs. (9) and (14). Neglecting higher-order terms again, the equation of motion is given by

$$\frac{d}{dt}\langle\sigma_{ga}(t)\sigma_{gb}(t')\rangle^{\mu\nu} = -i\tilde{\Omega}\langle\sigma_{ga}(t)\sigma_{gb}(t')\rangle^{\mu\nu} + \sqrt{\Gamma}\delta(t+r_1') \times\langle\sigma_{gb}(t')\rangle^{\nu}, \tag{22}$$

with the initial condition $\langle\sigma_{ga}(t')\sigma_{gb}(t')\rangle^{\mu\nu}=0$. From these equations, we have

$$G_{NL}(\mathbf{r},\mathbf{r}';t) = -\Gamma^2\prod_{j=1,2} e^{i\tilde{\Omega}(r_j-r_j'-t)}\theta(r_j'+t-R), \tag{23}$$

where $R=\mathrm{Max}(r_1,r_2)$. The step functions in Eq. (23) implies that the nonlinearity appear after both photons arrive at the atom. Thus, the two-photon propagator [Eqs. (15), (21), and (23)] is derived by solving the *semiclassical* equations of motion [Eqs. (20) and (22)].

C. Correlated input photons

We now observe the effects of correlation between two photons on the optical nonlinearity. For this purpose, we assume that the input wave function is given by the bivariate Gaussian,

$$f(r_1,r_2) = \frac{\exp\left[-\dfrac{\bar{r}_1^2+\bar{r}_2^2-2\rho\bar{r}_1\bar{r}_2}{4(1-\rho^2)d^2} + i\Omega(\bar{r}_1+\bar{r}_2)\right]}{(2\pi d^2)^{1/2}(1-\rho^2)^{1/4}}, \tag{24}$$

where $\bar{r}_j=r_j-a$. This wave function is characterized by three parameters, d (pulse length), ρ (correlation), and a (initial position). However, a is an irrelevant parameter satisfying $a<0$ and $|a|\ll d$. As indicated by the phase factor of $e^{i\Omega(r_1+r_2)}$, both input photons are resonant with the atom. It is of note that the single-photon profile, $I(r)=\int dr'|f(r,r')|^2 = (2\pi d^2)^{-1/2}\exp(-r^2/2d^2)$, is independent of ρ. The correlation parameter ρ lies in the $-1<\rho<1$ region, where positive and negative values indicate spatial correlation and anticorrelation, respectively, of two photons, and $\rho=0$ implies no correlation. In other words, two photons tend to arrive at the atom simultaneously as ρ is increased.

D. Results

The wave function after the interaction is obtained by performing the integral in Eq. (5). In order to quantify the nonlinear effects appearing in this wave function, we consider, as a reference, the *linear* output wave function defined by

$$g^L(r_1,r_2;t) = \int_{-\infty}^{0} dr_1' dr_2' G_H(r_1,r_1';t) G_V(r_2,r_2';t) f(r_1',r_2'). \tag{25}$$

This output is expected when two photons interact with the three-level atom independently. As a measure of optical nonlinearity, we employ the overlap α between g^L and g, as given by

$$\alpha = \int dr_1 dr_2 [g^L(r_1,r_2;t)]^* g(r_1,r_2;t). \tag{26}$$

This quantity becomes independent of t sufficiently after the interaction. $|\alpha| \leq 1$ by definition, and the nonlinear effect is reflected in the deviation of α from unity. In particular, this nonlinearity is applicable to optical gates if $\alpha = -1$ is achieved. α becomes real if two input photons are resonant with the atom, as in the present case.

When the input pulse length d is on the order of Γ^{-1}, photons are absorbed efficiently by the atom and large nonlinear optical effects result. In Fig. 3, fixing d at $2.5\Gamma^{-1}$, the dependence of α on the correlation parameter ρ is plotted. It is observed that the nonlinearity $|\alpha-1|$ becomes larger as ρ increases. This tendency can be easily understood, since nonlinear effects emerge only when the second photon arrives at the atom while the atom is excited by the first photon. The large nonlinearity observed here ($\alpha \simeq -0.9$) can never be achieved by uncorrelated photons [15,16]. However, the nonlinearity decreases in the $\rho \to 1$ limit, in which two photons arrive at the atom exactly at the same moment. This fact can be understood by the following considerations: the nonlinearity appears if the atom is not in the ground state (namely, excited by the first photon) when the second photon arrives at the atom. Therefore, a slight delay time between two photons is necessary for inducing nonlinear effects.

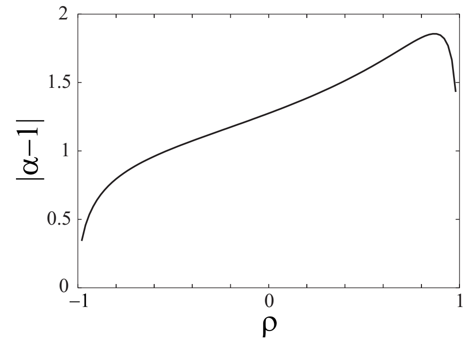

FIG. 3. Plot of $|\alpha-1|$ as a function of ρ. The pulse length is fixed at $d=2.5\Gamma^{-1}$. Maximum nonlinearity ($\alpha=-0.86$) is attained at $\rho=0.88$.

IV. CONCLUDING REMARKS

Two final remarks are in order. (i) The method proposed in this study is rigorous at the stage of solving the Scrhödinger equation in Eq. (2). However, in the Hamiltonian in Eq. (8), a rotating-wave approximation is employed and absence of pure dephasing is assumed. The effects of pure dephasing would become significant when the atom is embedded in a solid-state circumstance, such as a semiconductor quantum dot. (ii) Throughout this study, we assumed that the number of photons is conserved after the interaction. However, it is straightforward to extend the present method to cases in which the number of photons is not conserved. For example, for up-conversion of two H polarized photons into a V polarized photon, the propagator is obtained by $G(r,r_1',r_2';t) = \langle b_r(t)\rangle^{\mu\nu}$, where the average is taken over $|\phi\rangle = \mathcal{N}\exp(\mu a^\dagger_{r_1'} + \nu a^\dagger_{r_2'})$. Thus, the present method is applicable widely for the analysis of the coherent dynamics of photons.

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