Novel Method for Solving the Quantum Nonlinear Dynamics of Photons: Use of a Classical Input

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In order to investigate the nonlinear dynamics of photons, one must in principle solve a quantum many-particle problem, which usually requires intensive computation. In this study, we show that the spatial wave function of photons after nonlinear interaction can be obtained with less computation by assuming a classical input pulse and calculating a correlation function in the output field. This method is particularly useful when nonlinear optical media have many mechanical degrees of freedom, where quantum many-particle problems become extremely difficult.

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There has been rapid improvement in experimental techniques for generating and detecting photons in the number states recently [1–10]. Since photons can maintain quantum coherence for a long time, they are regarded as a promising candidate for implementing qubits. However, from the viewpoint of quantum computation, photonic qubits have the shortcoming that control of a photon by another photon is hard to realize, since the Kerr interaction between two photons is generally extremely weak at the single-photon level. However, there are several possibilities for overcoming this problem [11,12]. The optical nonlinearity of few-level quantum systems is expected to be sensitive to individual photons [13,14]. Recently, it has been suggested that even extremely weak Kerr nonlinearities can be utilized in quantum information processing, with the assistance of strong classical light fields [15]. In order to examine these possibilities and to design nonlinear photonic devices, quantitative analyses of the nonlinear dynamics of two photons (and of a photon and a classical light pulse) have become an important theoretical problem in the field of quantum optics [16,17].

A quantitative theory of two-photon nonlinear dynamics must satisfy the following two requirements. (i) Since quantum mechanics is required in order to describe photons in the number states, quantization of the photon field as well as the material system is essential [18,19]. (ii) Since nonlinear optical processes are generally sensitive to the spatial distribution of the photon field, the spatial profile of the photonic pulses should be characterized. For this purpose, the photon field must be treated rigorously as a multimode field. However, such rigorous analyses usually involve heavy computation and are practically impossible to perform for realistic material systems having many mechanical degrees of freedom. For this reason, multimode analyses of two-photon nonlinear dynamics have been performed only for simple nonlinear media, such as a two-level atom in a cavity [20–22].

In contrast, theories for treating the cases of classical input have been developed intensively [23,24]. [In this study, we use the term *classical* to indicate a field in the coherent state as given by Eq. (3), even when its amplitude is not large.] Since a classical field contains a two-photon component, it is natural to expect that traces of two-photon dynamics exist in the output field for a classical input [25]. From this perspective, we present in this study a method for obtaining full information of the two-photon dynamics, namely, the spatial wave function of the output photons, by considering a classical input and calculating a two-point correlation function of the output field. This method enables us to calculate the output wave function with less computation and is applicable even for cases in which the nonlinear medium has many mechanical degrees of freedom. Furthermore, extension of this method to the multiphoton cases is straightforward. Therefore, this method will serve as an effective theoretical tool for many problems in quantum nonlinear optics.

The problem to be considered in this study is stated as follows (see Fig. 1). Throughout this study, we denote the time variable by τ and set the initial and final moments at $\tau = 0$ and t, respectively. At the initial moment, two photons having the mode functions $f_a(r)$ and $f_b(r)$, normalized as $\int dr|f_a(r)|^2 = \int dr|f_b(r)|^2 = 1$ and localized in the $r<0$ region, are input into a nonlinear optical system. The overlap $\mathcal{V}$ between the two mode functions is given by $\mathcal{V} = \int dr f_a(r) f_b^*(r)$. Denoting the photon annihilation operator at the space coordinate r by c_r, the input state vector $|AB_{\rm in}\rangle$ is given by

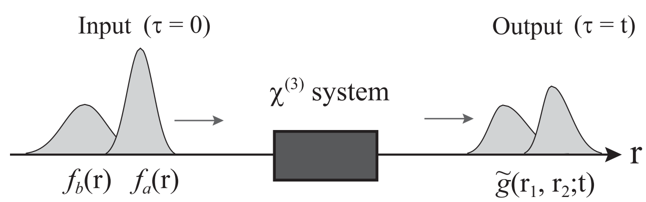

FIG. 1. Schematic illustration of the situation considered in this study. Two photons having the mode functions $f_a(r)$ and $f_b(r)$ are input into a third-order nonlinear optical system. The objective is to calculate the output wave function, $\tilde{g}(r_1, r_2; t)$.

$$|AB_{\rm in}\rangle = (1+|\mathcal{V}|^2)^{-1/2}\int dr_1 dr_2 f_a(r_1) f_b(r_2) c_{r_1}^\dagger c_{r_2}^\dagger |0\rangle. \tag{1}$$

The nonlinear optical medium is assumed to be a Kerr type and therefore the number of photons is conserved. Then, two photons appear in the output port after the nonlinear interaction in the medium. The output state vector $|AB_{\rm out}\rangle$ at the final moment can be written as

$$|AB_{\rm out}\rangle = 2^{-1/2}\int dr_1 dr_2 \tilde{g}(r_1, r_2; t) c_{r_1}^\dagger c_{r_2}^\dagger |0\rangle, \tag{2}$$

where the output wave function $\tilde{g}(r_1, r_2; t)$ is symmetrized as $\tilde{g}(r_1, r_2; t) = \tilde{g}(r_2, r_1; t)$ and is normalized as $\int dr_1 dr_2 |\tilde{g}(r_1, r_2; t)|^2 = 1$. In general, the output wave function $\tilde{g}$ is not factorizable after nonlinear interaction between two photons. The objective is to determine the output wave function $\tilde{g}(r_1, r_2; t)$ as a function of the input mode functions, $f_a(r)$ and $f_b(r)$. Since the input and output states are related through quantum time evolution by $|AB_{\rm out}\rangle = e^{-i\mathcal{H}t}|AB_{\rm in}\rangle$, where $\mathcal{H}$ is the Hamiltonian of the overall system composed of the photon field and the optical medium, a straightforward method to obtain $\tilde{g}(r_1, r_2; t)$ is to solve the quantum time evolution of the whole system. However, such a quantum many-particle problem is generally a computationally intensive task and at times it cannot be solved numerically, particularly when the optical medium has many mechanical degrees of freedom.

Instead of solving the dynamics of the two photons directly, we here consider the dynamics of a classical light pulse, the amplitude of which is given by $\mu f_a(r) + \nu f_b(r)$, where μ and ν are perturbation coefficients. The state vector corresponding to this classical pulse is given by

$$|\Phi_{\rm in}\rangle = \mathcal{N}\exp\left(\int dr[\mu f_a(r) + \nu f_b(r)]c_r^\dagger\right)|0\rangle, \tag{3}$$

where the normalization constant $\mathcal{N}$ is given by $\mathcal{N} = \exp[-2^{-1}(|\mu|^2 + |\nu|^2 + \mu\nu^*\mathcal{V} + \mu^*\nu\mathcal{V}^*)]$. Up to two-photon components, $|\Phi_{\rm in}\rangle$ can be expanded as

$$|\Phi_{\rm in}\rangle = \mathcal{N}[|0\rangle + \mu|A_{\rm in}\rangle + \nu|B_{\rm in}\rangle + 2^{-1/2}\mu^2|AA_{\rm in}\rangle + 2^{-1/2}\nu^2|BB_{\rm in}\rangle + (1+|\mathcal{V}|^2)^{1/2}\mu\nu|AB_{\rm in}\rangle], \tag{4}$$

where $|A_{\rm in}\rangle$ and $|AA_{\rm in}\rangle$ are the one- and two-photon states for the mode f_a, which are given by $|A_{\rm in}\rangle = \int dr f_a(r) c_r^\dagger|0\rangle$ and $|AA_{\rm in}\rangle = 2^{-1/2}[\int dr f_a(r) c_r^\dagger]^2|0\rangle$, respectively. $|B_{\rm in}\rangle$ and $|BB_{\rm in}\rangle$ are given similarly. The output state vector $|\Phi_{\rm out}\rangle$ for this classical input is given, as a result of quantum time evolution, by $|\Phi_{\rm out}\rangle = e^{-i\mathcal{H}t}|\Phi_{\rm in}\rangle$. Using the linearity of quantum time evolution and the photon-number conservation in Kerr systems, $|\Phi_{\rm out}\rangle$ is given by

$$|\Phi_{\rm out}\rangle = \mathcal{N}[|0\rangle + \mu|A_{\rm out}\rangle + \nu|B_{\rm out}\rangle + 2^{-1/2}\mu^2|AA_{\rm out}\rangle + 2^{-1/2}\nu^2|BB_{\rm out}\rangle + (1+|\mathcal{V}|^2)^{1/2}\mu\nu|AB_{\rm out}\rangle]. \tag{5}$$

The two-point correlation function for this output state is denoted by $\mathcal{G}(r_1, r_2; t) = \langle\Phi_{\rm out}|c_{r_1}c_{r_2}|\Phi_{\rm out}\rangle$. It is readily confirmed that the output wave function $\tilde{g}(r_1, r_2; t)$ is related to the second-order component of $\mathcal{G}$ proportional to $\mu\nu$ [hereafter denoted by $\mathcal{G}^{\mu\nu}(r_1, r_2; t)$] through the following relation:

$$\tilde{g}(r_1, r_2; t) = (2 + 2|\mathcal{V}|^2)^{-1/2}\mathcal{G}^{\mu\nu}(r_1, r_2; t). \tag{6}$$

Namely, we can determine the two-photon output function $\tilde{g}(r_1, r_2; t)$ by assuming a classical input pulse and calculating the two-point correlation function of the output field. This kind of problem has been investigated intensively in the context of squeezing, and theoretical prescriptions for this problem have been established [23,24]. As will be observed later, the principal task in the calculation of the two-point correlation function is to solve the differential equations for up to the second-order expectation values of the material system. This is obviously much simpler than solving a quantum many-particle problem and is particularly effective when the material system has many mechanical degrees of freedom, because quantum many-particle problems become extremely difficult in such cases.

In the following, we apply this method to a specific nonlinear optical system and calculate the two-photon output wave function. As a model nonlinear system, we employ a one-dimensional atom, which is realized in the weak coupling ($\kappa \gg g$) and dissipationless ($\gamma \to 0$) limit of a cavity quantum electrodynamics system [13,22]. Putting $\hbar = c = 1$, denoting the Pauli destruction operator of the atomic excitation by σ, and choosing the resonance of the atom as the origin of the energy (i.e., working in the rotating frame), the Hamiltonian of the overall system including the photon field and the optical system is given by

$$\mathcal{H} = \int dk[k c_k^\dagger c_k + \sqrt{\Gamma/2\pi}(c_k^\dagger\sigma + \sigma^\dagger c_k)], \tag{7}$$

where c_k is the photon annihilation operator in the wave number representation and Γ is the atomic decay rate. (Note that the cavity-field operator is eliminated adiabatically, and $\Gamma = 4g^2/\kappa$ in the language of cavity quantum electrodynamics.) Since we are considering the case of a classical input, the initial state vector is $|\Phi_{\rm in}\rangle$ as given by Eq. (3).

The Heisenberg equation for the atomic operator σ is given by [18,19]

$$\frac{d}{d\tau}\sigma = -\frac{\Gamma}{2}\sigma - i\sqrt{\Gamma}(1 - 2\sigma^\dagger\sigma)c_{-\tau}(0), \tag{8}$$

where $c_{-\tau}(0)$ is the initial annihilation operator for the photon field at $r = -\tau$. The field operator at the final moment t is given by

$$c_r(t) = c_{r-t}(0) - i\sqrt{\Gamma}\sigma(t-r), \quad (9)$$

which is known as the input-output relation [18,19]. It is readily confirmed that the output-field operators satisfy a proper commutation relation, $[c_r(t), c_{r'}^\dagger(t)] = \delta(r-r')$ [26]. Using the notation $\langle A\rangle \equiv \langle\Phi_{\rm in}|A|\Phi_{\rm in}\rangle$, the two-point correlation function in the output field is given by $\mathcal{G}(r_1, r_2; t) = \langle c_{r_1}(t)c_{r_2}(t)\rangle$. Since $\mathcal{G}(r_1, r_2; t) = \mathcal{G}(r_2, r_1; t)$ by definition, we take $r_1 < r_2$ in the following. Remembering that $c_r(0)$ and $\sigma(\tau)$ are commutable if $r + \tau < 0$ and that $|\Phi_{\rm in}\rangle$ is an eigenstate of the initial field operator $c_r(0)$, we obtain

$$\begin{aligned}\mathcal{G}(r_1, r_2; t) = \langle c_{r_1-t}(0)\rangle\langle c_{r_2-t}(0)\rangle \\ - i\sqrt{\Gamma}\langle\sigma(t-r_1)\rangle\langle c_{r_2-t}(0)\rangle \\ - i\sqrt{\Gamma}\langle\sigma(t-r_2)\rangle\langle c_{r_1-t}(0)\rangle \\ - \Gamma\langle\sigma(t-r_1)\sigma(t-r_2)\rangle. \quad (10)\end{aligned}$$

Note that $\langle c_r(0)\rangle$ is a known quantity, namely, the amplitude of the input field, $\mu f_a(r) + \nu f_b(r)$. Therefore, our task is to calculate the one- and two-time correlation functions, $\langle\sigma(\tau)\rangle$ and $\langle\sigma(\tau)\sigma(\tau')\rangle$. In order to obtain $\mathcal{G}^{\mu\nu}(r_1, r_2; t)$, the following three quantities are relevant: $x(\tau)$ [the component of $\langle\sigma(\tau)\rangle$ proportional to μ], $y(\tau)$ [the component of $\langle\sigma(\tau)\rangle$ proportional to ν], and $z(\tau, \tau')$ [the component of $\langle\sigma(\tau)\sigma(\tau')\rangle$ proportional to $\mu\nu$]. The equation of motion for $x(\tau)$ is given by

$$\frac{dx}{d\tau} = -\frac{\Gamma}{2}x - i\sqrt{\Gamma}f_a(-\tau), \quad (11)$$

with the initial condition of $x(0) = 0$. The equation of motion for $z(\tau, \tau')$ $(\tau > \tau')$ is given by

$$\frac{\partial z}{\partial\tau} = -\frac{\Gamma}{2}z - i\sqrt{\Gamma}[y(\tau')f_a(-\tau) + x(\tau')f_b(-\tau)], \quad (12)$$

with the initial condition of $z(\tau', \tau') = 0$. Equations (11) and (12) are integrated to give

$$x(\tau) = -i\sqrt{\Gamma}\int_{-\tau}^{\infty} d\mu f_a(\mu)e^{-\Gamma(\mu+\tau)/2}, \quad (13)$$

$$z(\tau, \tau') = x(\tau)y(\tau') + x(\tau')y(\tau) - 2x(\tau')y(\tau')e^{\Gamma(\tau'-\tau)/2}, \quad (14)$$

and $y(\tau)$ is obtained by replacing f_a with f_b in Eq. (13). Picking up the components proportional to $\mu\nu$ in Eq. (10) and using Eq. (6), the output wave function $\tilde{g}(r_1, r_2; t)$ is given by

$$\tilde{g}(r_1, r_2; t) = \tilde{g}_L(r_1, r_2; t) + \tilde{g}_{NL}(r_1, r_2; t) \quad (15)$$

$$\tilde{g}_L(r_1, r_2; t) = \frac{\bar{f}_a(r_1;t)\bar{f}_b(r_2;t) + \bar{f}_b(r_1;t)\bar{f}_a(r_2;t)}{\sqrt{2+2|\mathcal{V}|^2}}, \quad (16)$$

$$\tilde{g}_{NL}(r_1, r_2; t) = -\frac{2\Gamma^2}{\sqrt{2+2|\mathcal{V}|^2}}\iint_{\max(r_1,r_2)}^{\infty} d\tau_1 d\tau_2 f_a(\tau_1 - t) \times f_b(\tau_2 - t)e^{\Gamma(r_1+r_2-\tau_1-\tau_2)/2}, \quad (17)$$

where $\bar{f}_a$ is the one-photon output wave function, which is obtained as a result of a single-photon input in the mode f_a. It is given by

$$\bar{f}_a(r;t) = f_a(r-t) - \Gamma\int_r^{\infty} d\tau f_a(\tau - t)e^{\Gamma(r-\tau)/2}. \quad (18)$$

$\bar{f}_b$ is obtained by replacing f_a with f_b. Reflecting the fact that the output photons simply propagate at the light velocity sufficiently after the interaction, $\tilde{g}$ is a function of $r_1 - t$ and $r_2 - t$. Assuming that the two input photons have the same mode functions $[f_a = f_b = (2\Gamma^2/\pi)^{1/4} \times \exp(-\Gamma^2 r^2)]$, $\bar{f}_a$, $\tilde{g}_L$, and $\tilde{g}$ are visualized in Fig. 2.

Thus, we can obtain the two-photon output wave function $\tilde{g}$ by using a classical input. The practical task involves solving the time evolution of only three quantities. For this simple model system, we can determine the output wave function $\tilde{g}$ by directly solving the quantum mechanics for two quanta [20–22]. It is confirmed that $\tilde{g}$ thus calculated agrees with the current result, which demonstrates the validity of the proposed method.

Three comments should be made regarding the extensibility of the proposed method. (i) Derivation of Eq. (6) relies only on a fundamental property of quantum mechanics, namely, linearity of quantum time evolution. Thus, Eq. (6) is applicable not merely to photons but it can be applied to other elementary particles. Thus, if two-point correlation functions can be calculated easily for some particle, the proposed method may work as an effective tool for obtaining the two-body wave function after interaction. (ii) Let us briefly see the theoretical tasks required

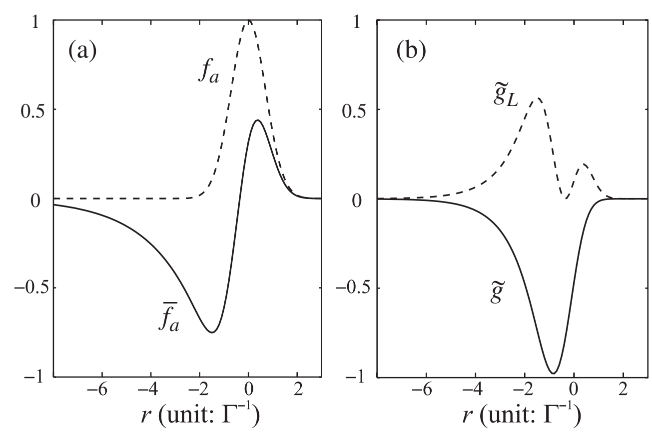

FIG. 2. (a) Plots of f_a (dotted line) and $\bar{f}_a$ (solid line). The unit of the vertical axis is $(2\Gamma^2/\pi)^{1/4}$. The origin of the horizontal axis is chosen at the center of f_a, and $\bar{f}_a$ is observed from the coordinate system moving at the light velocity. (b) Plots of $\tilde{g}$ (solid line) and $\tilde{g}_L$ (dotted line) on the diagonal line ($r = r'$). The unit of the vertical axis is $(2\Gamma^2/\pi)^{1/2}$.

when the optical medium has many mechanical degrees of freedom. As an example, we take a Frenkel excitonic system (composed of N two-level sites) placed inside a cavity [27]. Denoting the Pauli destruction operator for the jth site by σ_j and the annihilation operator for the cavity photon by a, the following quantities are relevant for calculating the two-point correlation function: $\langle a(t)\rangle$, $\langle \sigma_j(t)\rangle$, $\langle a(t)a(t)\rangle$, $\langle \sigma_j(t)a(t)\rangle$, $\langle \sigma_j(t)\sigma_i(t)\rangle$, $\langle a(t)a(t')\rangle$, and $\langle \sigma_j(t)a(t')\rangle$. Thus, when the mechanical degrees of freedom of the material system is N, the number of relevant expectation values is of the order of N^2. This is a much simpler numerical task than a quantum many-particle problem, in which, roughly speaking, the matrix size becomes of the order of N^2. Therefore, the present method is particularly effective when the optical medium has many mechanical degrees of freedom. (iii) Throughout this Letter, we are concerned with the two-photon case. However, it is straightforward to generalize the theory to cases in which more photons are involved. For example, in the case of three input photons having the mode functions f_a, f_b, and f_c, we should assume a classical pulse having the amplitude of $\mu f_a(r) + \nu f_b(r) + \xi f_c(r)$ and calculate the third-order component (proportional to $\mu\nu\xi$) of the three-point correlation function. The number of relevant expectation values is of the order of N^3, where N is the degree of freedom of the nonlinear medium. This is apparently simpler than solving the quantum mechanics of three quanta.

In summary, we have proposed an effective theoretical method for investigating the nonlinear dynamics of photons. This method enables us to calculate the output wave function $\tilde{g}(r_1, r_2)$ from the input mode functions $f_a(r)$ and $f_b(r)$, with far less computation than for quantum many-particle problems. The prescription is as follows: assume a classical light pulse, the amplitude of which is given by $\mu f_a(r) + \nu f_b(r)$, and calculate the two-point correlation function $\mathcal{G}^{\mu\nu}(r_1, r_2)$ of the output field; then, the output two-photon wave function is given by Eq. (6). The validity and the simplicity of the method is demonstrated by taking a one-dimensional atom as a model nonlinear system, where the output wave function can be obtained by solving the time evolution of only three quantities. This method is particularly effective when the nonlinear medium has many mechanical degrees of freedom and when many photons are involved, because quantum many-particle problems become extremely difficult in such cases. Furthermore, since the derivation of Eq. (6) relies only on the linearity of quantum time evolution, it would be applicable to general elementary particles other than photons. Thus, the method proposed here would be useful for many problems in quantum optics, particularly in designing optical quantum information devices, where quantitative consideration of the fidelity between photonic pulses is essential.

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