Evaluation of multiphoton effects in down-conversion

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(Received 28 January 2010; published 7 April 2010)

Multiphoton effects in down-conversion are investigated based on the full-quantum multimode formalism by considering a three-level system as a prototype nonlinear system. We analytically derive the three-photon output wave function for two input photons, where one of the two input photons is down-converted and the other one is not. Using this output wave function, we calculate the down-conversion probability, the purity, and the fidelity to evaluate the entanglement between a down-converted photon pair and a non-down-converted photon. It is shown that the saturation effect occurs by multiphoton input and that it affects both the down-conversion probability and the quantum correlation between the down-converted photon pair and the non-down-converted photon. We also reveal the necessary conditions for multiphoton effects to be strong.

DOI: 10.1103/PhysRevA.81.043806 PACS number(s): 42.65.Lm, 42.65.Sf

I. INTRODUCTION

Parametric down-conversion is a nonlinear optical process in which a parent photon is converted into two daughter photons; this process satisfies energy and momentum conservation [1,2]. Parametric down-conversion has recently been attracting considerable interest in the context of fundamental tests of quantum mechanics and of quantum-information processing, since it is the most efficient and convenient method for generating polarization entanglement between two particles [3–5].

As a classical nonlinear optical process, parametric down-conversion has been explained in terms of coupled-mode theory, which is based on the linear and nonlinear susceptibilities of optical media [6–8]. The concept of several light modes interacting through the nonlinear susceptibility is also useful when constructing a quantum-mechanical theory of down-conversion. The simplest phenomenological interaction Hamiltonian is the following [9,10]:

$$\mathcal{H} = \lambda a_s^\dagger a_i^\dagger a_p + \text{H.c.}, \tag{1}$$

where a_s, a_i, and a_p are, respectively, the annihilation operators for the signal, idler, and pump modes and λ is a coupling constant that is proportional to the nonlinear susceptibility and the interaction time (crystal length). While such phenomenological theories can describe several basic properties of the process, they have the following two drawbacks: (i) They treat signal and idler photons as independent particles. However, in actual down-conversion, the signal and idler photons are strongly correlated in time. (ii) They assume that input pump photons are down-converted independently. Specifically, when n photons are input into a nonlinear media and only one photon is down-converted, its dynamics is unaffected by the other $n-1$ photons. However, in principle, the $n-1$ pump photons and the down-converted photons are spatiotemporally correlated after nonlinear interaction. When down-converted photons are used as a quantum-mechanical light source, it is essential to rigorously characterize the down-converted photons, including their spatiotemporal properties.

The purpose of this study is to investigate multiphoton effects in down-conversion based on the full-quantum multimode formalism, in which both the photon field and the atomic system are quantized and the multimode nature of the photon field is exactly taken into account. Taking a three-level system as a prototype system for a nonlinear medium, we formulate an analytical method for obtaining various quantities characterizing multiphoton effects such as the probability, the purity, and the fidelity. We clarify that the nonlinearity induced by saturation of the three-level system affects both the probability and the quantum correlation between down-converted photons and a fundamental (*non*-down-converted) photon.

II. FORMULATION

A. Hamiltonian

In this study, we consider a physical setup in which photons propagating in one dimension are down-converted at a three-level system (hereafter, atom) located at $r=0$, as illustrated in Fig. 1 [11–13]. The atom has three levels: $|g\rangle$, $|m\rangle$, and $|e\rangle$. The energies of $|e\rangle$ and $|m\rangle$ relative to that of $|g\rangle$ are denoted by Ω_e and Ω_m, respectively. The $|e\rangle \leftrightarrow |g\rangle$ transition is assisted by a horizontally polarized photon, whereas $|e\rangle \leftrightarrow |m\rangle$ and $|m\rangle \leftrightarrow |g\rangle$ transitions are assisted by vertically polarized photons; the spontaneous decay rates for the $|e\rangle \to |g\rangle$, $|e\rangle \to |m\rangle$, and $|m\rangle \to |g\rangle$ transitions are denoted by Γ_1, Γ_2, and Γ_3, respectively. Under the rotating wave approximation, the Hamiltonian for the overall system is given by (setting $\hbar = c = 1$)

$$\begin{aligned}\mathcal{H} = {}& \Omega_e\sigma_{ee} + \Omega_m\sigma_{mm} + \int dk k h_k^\dagger h_k + \int dk k v_k^\dagger v_k \\ & + (i\sqrt{\Gamma_1}\sigma_{eg}\tilde{h}_{r=0} + i\sqrt{\Gamma_2}\sigma_{em}\tilde{v}_{r=0} \\ & + i\sqrt{\Gamma_3}\sigma_{mg}\tilde{v}_{r=0} + \text{H.c.}),\end{aligned} \tag{2}$$

where σ_{ij} $(=|i\rangle\langle j|)$ is the atomic transition operator and h_k (v_k) is the annihilation operator for the horizontally (vertically) polarized photon with wave number k. The

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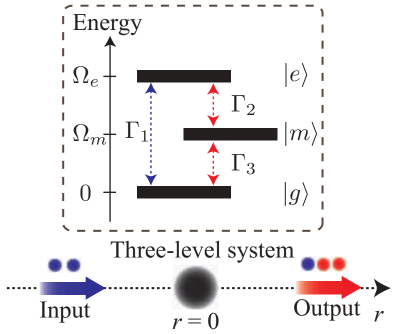

FIG. 1. (Color online) Schematic illustration of the system considered in this study. Two photons resonant with the $|g\rangle \to |e\rangle$ transition are input into a three-level atom. On interacting with the atom, two down-converted photons are generated with some probability.

real-space representations $\tilde{h}_r$ and $\tilde{v}_r$ are the Fourier transforms of h_k and v_k, respectively: $\tilde{h}_r = (2\pi)^{-1/2}\int dk e^{ikr} h_k$ and $\tilde{v}_r = (2\pi)^{-1/2}\int dk e^{ikr} v_k$.

B. Input and output states

To observe multiphoton effects that occur in down-conversion, we investigate the minimal case in which two photons are simultaneously input to the system. The initial atom-photon state vector is

$$\left|\psi_{\rm in}^{(2)}\right\rangle = \frac{1}{\sqrt{2}}\left[\int dr f(r)\tilde{h}_r^\dagger\right]^2 |0\rangle, \tag{3}$$

where $|0\rangle$ represents the overall ground state (the product of the atomic ground state and the photonic vacuum state). Note that the two input photons are uncorrelated and have the same wave function $f(r)$ as photons in classical pump pulses. $f(r)$ is normalized as $\int dr|f(r)|^2 = 1$ and localized in the input port ($r < 0$ region). After interacting with the atom, down-converted photons may be generated due to the cascade decay channel of $|e\rangle \to |m\rangle \to |g\rangle$. The output state vector can be formally written as

$$\begin{aligned}\left|\psi_{\rm out}^{(2)}\right\rangle = &\int d^2r \frac{g_{2\to 2}(r_1,r_2;t)}{\sqrt{2}}\tilde{h}_{r_1}^\dagger \tilde{h}_{r_2}^\dagger|0\rangle \\ &+ \int d^3r \frac{g_{2\to 3}(r_1,r_2,r_3;t)}{\sqrt{2}}\tilde{h}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger \tilde{v}_{r_3}^\dagger|0\rangle \\ &+ \int d^4r \frac{g_{2\to 4}(r_1,r_2,r_3,r_4;t)}{\sqrt{4!}}\tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger \tilde{v}_{r_3}^\dagger \tilde{v}_{r_4}^\dagger|0\rangle, \end{aligned}\tag{4}$$

where t is the time at which the atom is completely deexcited, and $g_{2\to j}$ represents the wave function of the j output photons ($4-j$ fundamental photons and $2j-4$ down-converted ones). The output wave functions satisfy the sum rule of $\sum_{j=2}^4(\int d^jr|g_{2\to j}|^2) = 1$ since $\langle\psi_{\rm out}^{(2)}|\psi_{\rm out}^{(2)}\rangle = 1$.

To quantify multiphoton effects, we need to consider a reference case in which each photon is down-converted independently without being affected by the other photons. For this purpose, we consider the input of a single photon and its resultant output:

$$\left|\psi_{\rm in}^{(1)}\right\rangle = \int dr f(r)\tilde{h}_r^\dagger|0\rangle, \tag{5}$$

$$\begin{aligned}\left|\psi_{\rm out}^{(1)}\right\rangle = &\int dr g_{1\to 1}(r;t)\tilde{h}_r^\dagger|0\rangle \\ &+ \int dr_1 dr_2 \frac{g_{1\to 2}(r_1,r_2;t)}{\sqrt{2}}\tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger|0\rangle, \end{aligned}\tag{6}$$

where $g_{1\to j}$ represents the wave function of j output photons ($2-j$ fundamental photons and $2j-2$ down-converted ones). The output wave functions satisfy the sum rule of $\sum_{j=1}^2(\int d^jr|g_{1\to j}|^2) = 1$.

C. Relation between input and output wave functions

The output state vector is determined by the Schrödinger equation

$$\left|\psi_{\rm out}^{(1),(2)}\right\rangle = e^{-i\mathcal{H}t}\left|\psi_{\rm in}^{(1),(2)}\right\rangle. \tag{7}$$

As demonstrated in the Appendix, this equation can be solved analytically by considering a coherent state as the input state [11,12]. Here, we summarize the results of the relation between the input and the output wave functions. The output wave functions for one-photon input ($g_{1\to 1}$ and $g_{1\to 2}$) are given by

$$g_{1\to 1}(r;t) = f(r-t) - \sqrt{\Gamma_1}\langle\sigma_{ge}(t-r)\rangle, \tag{8}$$

$$g_{1\to 2}(r_1,r_2;t) = \sqrt{\Gamma_2\Gamma_3}\langle\sigma_{gm}(t-r_1)\sigma_{me}(t-r_2)\rangle, \tag{9}$$

where $\langle\sigma_{ge}\rangle$ and $\langle\sigma_{me}\rangle$ are respectively the *linear* one- and two-time atomic correlation functions, which are given by

$$\langle\sigma_{ge}(t)\rangle = \sqrt{\Gamma_1}\int_0^\infty d\xi f(-t+\xi)e^{-{\rm i}\tilde{\Omega}_e\xi}, \tag{10}$$

$$\langle\sigma_{gm}(t_1)\sigma_{me}(t_2)\rangle = \langle\sigma_{ge}(t_l)\rangle e^{{\rm i}\tilde{\Omega}_m(t_l-t_s)}, \tag{11}$$

where $\tilde{\Omega}_e = \Omega_e - i(\Gamma_1+\Gamma_2)/2$, $\tilde{\Omega}_m = \Omega_m - {\rm i}\Gamma_3/2$, $t_l = \max(t_1,t_2)$, and $t_s = \min(t_1,t_2)$. The probabilities of the nonoccurrence and the occurrence of down-conversion are respectively given by $P_{1\to 1} = \int dr|g_{1\to 1}(r;t)|^2$ and $P_{1\to 2} = \int dr_1 dr_2|g_{1\to 2}(r_1,r_2;t)|^2$, which satisfy $P_{1\to 1}+P_{1\to 2} = 1$.

For the case of two-photon input, we present the three-photon output wave function, $g_{2\to 3}$. (See Appendix for $g_{2\to 2}$ and $g_{2\to 4}$.) It is given by

$$\begin{aligned}&g_{2\to 3}(r,r_1,r_2;t) \\ &\quad = g_{1\to 1}(r;t)g_{1\to 2}(r_1,r_2;t) + \delta g_{2\to 3}(r,r_1,r_2;t), \end{aligned}\tag{12}$$

where $\delta g_{2\to 3}(r,r_1,r_2;t)$ is given by

$$\delta g_{2\to 3}(r,r_1,r_2;t) = \begin{cases}\sqrt{\Gamma_1\Gamma_2\Gamma_3}\langle\sigma_{ge}(t-r_1)\rangle\langle\sigma_{gm}(t-r_1)\sigma_{me}(t-r_2)\rangle e^{-{\rm i}\tilde{\Omega}_e(r_1-r)} & (r \leqslant r_1 \leqslant r_2) \\ \sqrt{\Gamma_1\Gamma_2\Gamma_3}\langle\sigma_{ge}(t-r)\rangle\langle\sigma_{gm}(t-r)\sigma_{me}(t-r_2)\rangle e^{-{\rm i}\tilde{\Omega}_m(r-r_1)} & (r_1 \leqslant r \leqslant r_2) \\ \sqrt{\Gamma_1\Gamma_2\Gamma_3}\langle\sigma_{ge}(t-r)\rangle\langle\sigma_{gm}(t-r_2)\sigma_{me}(t-r_1)\rangle e^{-{\rm i}\tilde{\Omega}_e(r-r_2)} & (r_1 \leqslant r_2 \leqslant r).\end{cases} \tag{13}$$

The first term on the right-hand side of Eq. (12) is obtained when two input photons are down-converted independently of each other, and the second term corresponds to the correction for multiphoton effects. The probability for three-photon output is given by $P_{2\to 3} = \int dr dr_1 dr_2 |g_{2\to 3}(r, r_1, r_2; t)|^2$.

D. Measures of multiphoton effects

In this subsection, we focus on the case in which two photons are input simultaneously and only one of them is down-converted (namely, the three-photon component in the output). From Eq. (4), we introduce the following *target* state vector:

$$\left|\psi_{\rm out}^{2\to 3}\right\rangle \equiv \int d^3r \frac{g_{2\to 3}(r, r_1, r_2; t)}{\sqrt{2}} \tilde{h}_r^\dagger \tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger |0\rangle. \tag{14}$$

This state includes multiphoton effects. In contrast, assuming that each input photon is down-converted independently, we can construct the *reference* three-photon state from Eqs. (3) and (6). It is given by

$$\left|\widetilde{\psi}_{\rm out}^{2\to 3}\right\rangle \equiv \int d^3r g_{1\to 1}(r; t) g_{1\to 2}(r_1, r_2; t) \tilde{h}_r^\dagger \tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger |0\rangle. \tag{15}$$

Hereafter, quantities with tildes represent those concerning this reference state, which is free from multiphoton effects.

1. Down-conversion probability

The difference between the aforementioned two states will appear in the down-conversion probabilities, which are determined by the norms of the aforementioned state vectors:

$$P_{2\to 3} = {\rm Tr}\left[\left|\psi_{\rm out}^{2\to 3}\right\rangle\left\langle\psi_{\rm out}^{2\to 3}\right|\right] = \int d^3r |g_{2\to 3}(r, r_1, r_2; t)|^2, \tag{16}$$

$$\widetilde{P}_{2\to 3} = {\rm Tr}\left[\left|\widetilde{\psi}_{\rm out}^{2\to 3}\right\rangle\left\langle\widetilde{\psi}_{\rm out}^{2\to 3}\right|\right] = 2P_{1\to 1}P_{1\to 2}, \tag{17}$$

where Tr indicates the trace over all photons. Rigorously, $P_{2\to 3}$ and $\tilde{P}_{2\to 3}$ are functions of t; however, they almost become independent of t when sufficient time has passed and we take this limit in the rest of this article.

2. Purity

The density matrices of down-converted photons are obtained from the aforementioned state vectors by tracing out the fundamental photon:

$$\rho_{\rm dc} = \frac{{\rm Tr}_h\left[\left|\psi_{\rm out}^{2\to 3}\right\rangle\left\langle\psi_{\rm out}^{2\to 3}\right|\right]}{{\rm Tr}\left[\left|\psi_{\rm out}^{2\to 3}\right\rangle\left\langle\psi_{\rm out}^{2\to 3}\right|\right]} = \frac{\int d^4r \int d\xi\, g_{2\to 3}(\xi, r_1, r_2; t) g_{2\to 3}^*(\xi, r_3, r_4; t) \tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger |0\rangle\langle 0| \tilde{v}_{r_3}\tilde{v}_{r_4}}{2P_{2\to 3}}, \tag{18}$$

$$\widetilde{\rho}_{\rm dc} = \frac{{\rm Tr}_h\left[\left|\widetilde{\psi}_{\rm out}^{2\to 3}\right\rangle\left\langle\widetilde{\psi}_{\rm out}^{2\to 3}\right|\right]}{{\rm Tr}\left[\left|\widetilde{\psi}_{\rm out}^{2\to 3}\right\rangle\left\langle\widetilde{\psi}_{\rm out}^{2\to 3}\right|\right]} = \frac{\int d^4r g_{1\to 2}(r_1, r_2; t) g_{1\to 2}^*(r_3, r_4; t) \tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger |0\rangle\langle 0| \tilde{v}_{r_3}\tilde{v}_{r_4}}{2P_{1\to 2}}, \tag{19}$$

where ${\rm Tr}_h$ indicates a partial trace over only the horizontally polarized photon.

The purity $\mathcal{P}$ is a measure of the entanglement between the two down-converted photons and the residual fundamental photon. It is defined by

$$\mathcal{P} = {\rm Tr}_v\left(\rho_{\rm dc}^2\right), \tag{20}$$

$$\widetilde{\mathcal{P}} = {\rm Tr}_v\left(\tilde{\rho}_{\rm dc}^2\right) = 1, \tag{21}$$

where ${\rm Tr}_v$ represents the trace over the vertically polarized photons. By definition, $0 \leqslant \mathcal{P} \leqslant 1$ is satisfied. The down-converted photons are separable from the residual photon in the reference state of Eq. (15); thus $\widetilde{\mathcal{P}} = 1$. However, when several photons are input simultaneously, the down-converted photons may entangle with the fundamental photon, reducing $\mathcal{P}$. Using Eq. (18), $\mathcal{P}$ can be rewritten as

$$\mathcal{P} = \frac{\int d^4r \left|\int d\xi\, g_{2\to 3}(\xi, r_1, r_2; t) g_{2\to 3}^*(\xi, r_3, r_4; t)\right|^2}{P_{2\to 3}^2}. \tag{22}$$

3. Fidelity

The fidelity $\mathcal{F}$ is a measure of the closeness between $\rho_{\rm dc}$ and $\tilde{\rho}_{\rm dc}$ defined by

$$\mathcal{F} = [{\rm Tr}_v(\rho_{\rm dc}\tilde{\rho}_{\rm dc})]^{1/2}. \tag{23}$$

Note that $\mathcal{F}$ reaches 1 when $\rho_{\rm dc}$ is equal to $\tilde{\rho}_{\rm dc}$, while $\mathcal{F}$ decreases with increasing distance between the two states due to multiphoton effects. Using Eqs. (18) and (19), $\mathcal{F}$ can be rewritten as

$$\mathcal{F} = \left[\frac{\int d^4r \int d\xi\, g_{1\to 2}^*(r_1, r_2) g_{2\to 3}(\xi, r_1, r_2; t) g_{2\to 3}^*(\xi, r_1', r_2'; t) g_{1\to 2}(r_1' r_2')}{P_{1\to 2}P_{2\to 3}}\right]^{1/2}. \tag{24}$$

III. RESULTS

In this section, we present the numerical results assuming a specific form of the input photonic wave function $f(r)$. We employ the following form of $f(r)$:

$$f(r) = \begin{cases} (2/d)^{1/2} \exp(r/d + i\omega r) & (r < 0) \\ 0 & (r > 0) \end{cases}, \qquad (25)$$

where d is the pulse length and ω is the frequency of the input photon that is close to the $|g\rangle \to |e\rangle$ transition. Using Eq. (10), the linear response of the atom induced by this photon is given by

$$\langle \sigma_{ge}(t) \rangle = \begin{cases} 0 & (t < 0) \\ (2\Gamma_1/d)^{1/2}\left[e^{-(d^{-1}+i\omega)t} - e^{-i\tilde{\Omega}_e t}\right][-i\Delta + (\Gamma_1+\Gamma_2)/2 - d^{-1}]^{-1} & (t > 0), \end{cases} \qquad (26)$$

where $\Delta(=\omega - \Omega_e)$ is the detuning. For simplicity, we hereafter assume that $\Gamma_1 = \Gamma_2 = \Gamma_3$ and $\Omega_m = \Omega_e/2$.

A. Probability

1. One-photon input

We preliminarily observe the probabilities for the occurrence ($P_{1\to2}$) and nonoccurrence ($P_{1\to1}$) of down-conversion when only a single photon is input. In Fig. 2, $P_{1\to1}$ and $P_{1\to2}$ are plotted as functions of the pulse length d, assuming complete resonance ($\Delta = 0$). The down-conversion probability $P_{1\to2}$ is a monotonically increasing function of d. The down-conversion probability vanishes in the short-pulse limit ($d \ll \Gamma_1^{-1}$), since the input photon becomes spectrally broad in this limit and is no longer resonant with the atom. In contrast, the down-conversion probability becomes unity in the long-pulse limit ($d \gg \Gamma_1^{-1}$). The cause of this nearly perfect down-conversion is the destructive interference between the incident photon and the atomic radiation. Equation (8) shows that the output photon at the fundamental frequency is composed of the incident photon f and the atomic radiation $\langle \sigma_{ge} \rangle$; in the long-pulse limit, Eq. (10) can be adiabatically integrated to give $\langle \sigma_{ge}(t) \rangle = 2\sqrt{\Gamma_1} f(-t)/(\Gamma_1 + \Gamma_2)$, which completely cancels the incident photon f when $\Gamma_1 = \Gamma_2$.

When the photon frequency is detuned ($\Delta \neq 0$), this destructive interference becomes incomplete, and the probability of the down-conversion decreases. In Fig. 3, $P_{1\to1}$ and $P_{1\to2}$ are plotted as functions of the detuning Δ, fixing the pulse length at $d = 10\Gamma_1^{-1}$. The down-conversion probability decreases monotonically as the detuning $|\Delta|$ is increased.

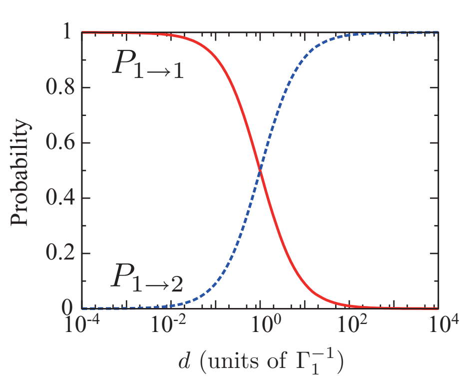

FIG. 2. (Color online) $P_{1\to1}$ (solid line) and $P_{1\to2}$ (dashed line) as functions of the pulse length d. The input pulse is in resonance with the $|g\rangle \to |e\rangle$ transition ($\Delta = 0$).

2. Two-photon input

Next, we investigate the probability for the case when two photons are input. We focus on the three-photon output state which is composed of a down-converted photon pair and a fundamental photon. Figure 4 shows the probability $P_{2\to3}$ at the resonance frequency ($\Delta = 0$) as a function of the pulse length d. It is observed that the probability vanishes in the short-pulse limit $d \ll \Gamma_1^{-1}$. As d increases, the down-conversion process becomes more dominant around $d \sim \Gamma_1^{-1}$ due to efficient excitation and emission of the atom, as found in the case of one-photon input. Consequently, $P_{2\to3}$ increases as d approaches $d \sim \Gamma_1^{-1}$. Since further increase in d make down-conversion more efficient, the process in which both input photons are down-converted into four photons becomes dominant. Therefore, $P_{2\to3}$ starts to decrease and approaches zero in the long-pulse limit $d \gg \Gamma_1^{-1}$.

To study the nonlinear effect, we need to compare the aforementioned result with the one for an *uncorrelated* three-photon output state where there is no correlation between the fundamental photon and the down-converted photons. The dashed line in Fig. 4 shows the probability $\tilde{P}_{2\to3}$. As expected from Fig. 2, $\tilde{P}_{2\to3}$ has a peak around $d \sim \Gamma_1^{-1}$. However, the peak height and the peak position are shifted from those for the probability $P_{2\to3}$ for the stimultaneous two-photon input. This difference is due to multiphoton effects, because the nonlinear effect does not occur for independent two-photon input.

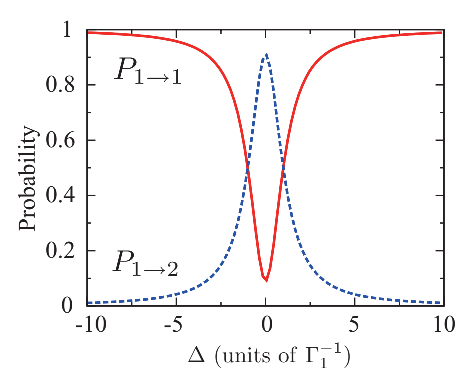

FIG. 3. (Color online) $P_{1\to1}$ (solid line) and $P_{1\to2}$ (dashed line) as functions of the detuning Δ. The pulse length is fixed at $d = 10\Gamma_1^{-1}$.

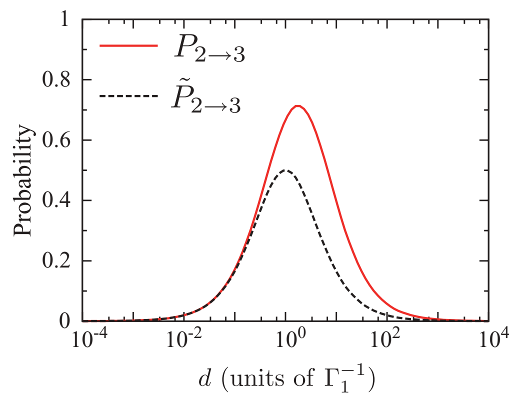

FIG. 4. (Color online) $P_{2\to3}$ (solid line) and $\tilde{P}_{2\to3}$ (dashed line) as functions of the pulse length d. The input photon is resonant with the $|g\rangle \to |e\rangle$ transition ($\Delta = 0$).

Multiphoton effects in the probability $P_{2\to3}$ can be discussed analytically by noting $P_{2\to3} = \int d^3r |g_{2\to3}|^2$ and $g_{2\to3} = g_{1\to1}g_{1\to2} + \delta g_{2\to3}$. Here, we find that $\delta g_{2\to3}$ represents multiphoton effects, since the probability $P_{2\to3}$ coincides with $\tilde{P}_{2\to3}$ for $\delta g_{2\to3} = 0$. The analytic form of $\delta g_{2\to3}$ given by Eq. (13) indicates that the nonlinearity is induced by multiple interference among the atomic transitions. For a short pulse ($d \ll \Gamma_1^{-1}$), down-converted photons are hardly produced, and consequently the nonlinear factor $\delta g_{2\to3}$ becomes almost zero. For a long pulse ($d \gg \Gamma_1^{-1}$), down-converted photons are efficiently produced, while the excited states of the atom quickly decay to the ground state compared with a typical time scale d, at which multiple interference becomes relevant. In this case, the nonlinear effect disappears since there is no chance of interference between the down-converted photons and the non-down-converted photon. The nonlinear effect (i.e., the difference between $P_{2\to3}$ and $\tilde{P}_{2\to3}$) is most significant near $d \sim \Gamma_1^{-1}$ as seen in Fig. 4. Compared with independent two-photon input, the peak of the probability $P_{2\to3}$ shifts to a larger pulse length d. This trend reflects the retarded nature of the nonlinear effect, which becomes effective only after a certain number of down-converted photons are generated. These features of stimultaneous two-photon input are expected to carry over to a general input of more than two correlated photons.

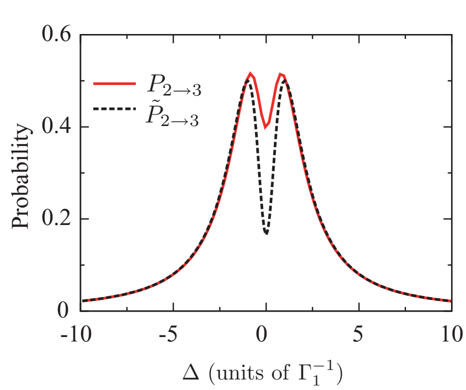

FIG. 5. (Color online) $P_{2\to3}$ (solid line) and $\tilde{P}_{2\to3}$ (dashed line) as functions of the detuning Δ. The pulse length is fixed at $d = 10\ \Gamma_1^{-1}$.

Multiphoton effects are most significant for the resonance condition $\Delta = 0$, at which excitation of the atom occurs efficiently. To see this, we plot $P_{2\to3}$ and $\tilde{P}_{2\to3}$ as functions of Δ in Fig. 5. The result for an uncorrelated three-photon output state can be understood by noting that $\tilde{P}_{2\to3} = 2P_{1\to2}P_{1\to1}$. The two peaks in this figure appear by multiplying the Δ dependences of $P_{1\to1}$ and $P_{1\to1}$ shown in Fig. 3. The difference between $P_{2\to3}$ and $\tilde{P}_{2\to3}$ indicates that the nonlinear effect is effective only near $\Delta = 0$.

B. Spatial profile of down-converted photons

Multiphoton effects almost appear in the spatial profile of down-converted photons. The spatial profiles are given by

$$I_{\rm out}^{1\to2}(r_1, r_2; t) = \frac{\langle \psi_{\rm out}^{1\to2} | \tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger \tilde{v}_{r_2} \tilde{v}_{r_1} | \psi_{\rm out}^{1\to2} \rangle}{P_{1\to2}} = \frac{|g_{1\to2}(r_1, r_2; t)|^2}{P_{1\to2}}, \tag{27}$$

$$I_{\rm out}^{2\to3}(r_1, r_2; t) = \frac{\langle \psi_{\rm out}^{2\to3} | \tilde{v}_{r_1}^\dagger \tilde{v}_{r_2}^\dagger \tilde{v}_{r_2} \tilde{v}_{r_1} | \psi_{\rm out}^{2\to3} \rangle}{P_{2\to3}} = \frac{\int d\xi\, |g_{2\to3}(\xi, r_1, r_2; t)|^2}{P_{2\to3}}, \tag{28}$$

for one and two input photons, respectively. Note that $I_{\rm out} \geqslant 0$, $\int d^2r\, I_{\rm out} = 1$, and $I_{\rm out}(r_1, r_2) = I_{\rm out}(r_2, r_1)$, by definition.

Figure 6 shows contour plots of $I_{\rm out}^{1\to2}$ and $I_{\rm out}^{2\to3}$. Both $I_{\rm out}^{1\to2}$ and $I_{\rm out}^{2\to3}$ are distributed near the diagonal line, indicating strong temporal entanglement, which is usually observed in down-converted photons. Comparison of $I_{\rm out}^{1\to2}$ and $I_{\rm out}^{2\to3}$ confirms that additional input photons reduce the temporal entanglement between the down-converted photons.

C. Purity and fidelity

In this subsection, we evaluate multiphoton effects in down-conversion by the purity $\mathcal{P}$ and the fidelity $\mathcal{F}$, which are given by Eqs. (22) and (24), respectively.

First, we examine the pulse length d dependences of $\mathcal{P}$ and $\mathcal{F}$ at $\Delta = 0$. Figure 7 shows that they decrease monotonically with increasing d. They reach finite values in the $d \gg \Gamma_1^{-1}$ region. Figure 8 shows the detuning Δ dependences of $\mathcal{P}$ and

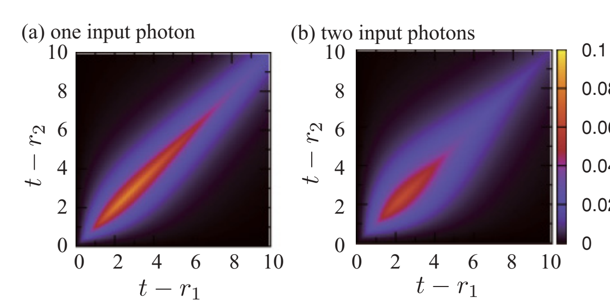

FIG. 6. (Color online) Spatial profiles of down-converted photons generated by (a) one input photon and (b) two input photons. The input photons are in resonance with the $|g\rangle \to |e\rangle$ transition ($\Delta = 0$). The pulse length is fixed at $d = 10\Gamma_1^{-1}$.

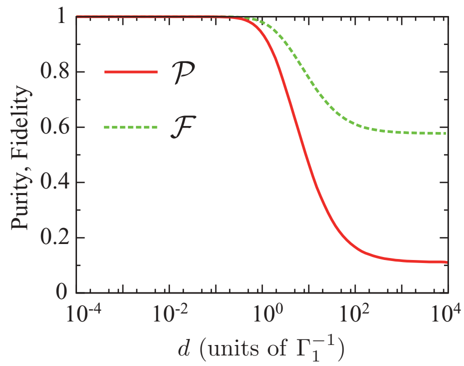

FIG. 7. (Color online) $\mathcal{P}$ and $\mathcal{F}$ as functions of the pulse length d. The input photon is in resonance with the $|g\rangle \to |e\rangle$ transition ($\Delta = 0$).

$\mathcal{F}$. They have a minimum value at the resonance frequency ($\Delta = 0$) and increase monotonically as functions of $|\Delta|$. These results indicate that a longer pulse and a smaller $|\Delta|$ are advantageous for generating entanglement between the residual fundamental photon and the down-converted photon pair.

We briefly discuss the mechanism of the behaviors of $\mathcal{P}$ and $\mathcal{F}$. When $\delta g_{2\to3}$ vanishes, Eqs. (12), (22), and (24) show that both the purity and the fidelity approach 1. Therefore, $\delta g_{2\to3}$ has large effects on both $\mathcal{P}$ and $\mathcal{F}$. As discussed in the previous section, this term depends on the atomic emission $\langle\sigma_{ge}\rangle$. When d is sufficiently short or $|\Delta|$ is large, the first excitation is strongly suppressed and $\langle\sigma_{ge}\rangle$ approaches 0. In other words, there are too few generated down-converted photon pairs to produce multiphoton effects in these limits; thus, the fundamental photon and the down-converted photon pair develop independently. Therefore, both the purity and the fidelity become 1. On the other hand, in the $d \gg \Gamma_1^{-1}$ region, atomic emission becomes large (as discussed for the one-input-photon case) and both the purity and the fidelity for the three-photon state remain at the resonance frequency.

IV. SUMMARY

In this study, we have investigated multiphoton effects in down-conversion in a three-level system based on the full-quantum multimode formalism. We have analytically derived the three-photon output wave function for two input photons and shown that the saturation effect for multiphoton input occurs and the correlation strength is weak due to the spatial shape of down-converted photons. We also introduce the purity and the fidelity to determine the quantum state for three-photon output. At the resonance frequency $\Delta = 0$, we demonstrated that the correlation between the transmitted output photon and a down-converted photon pair is weak when the pulse length d is sufficiently short, whereas the correlation increases greatly around $d \approx \Gamma_1^{-1}$ due to an increase in the number of down-converted photons. The correlation remains when photons are down-converted with almost unit efficiency. This situation is realized under the following conditions: the $|e\rangle \to |g\rangle$ and $|e\rangle \to |m\rangle$ transitions have similar decay rates and the input photons are resonant with the $|g\rangle \to |e\rangle$ transition. Under these conditions, the nonlinear term is comparable to the linear term and entanglement remains even when the pulse length is long.

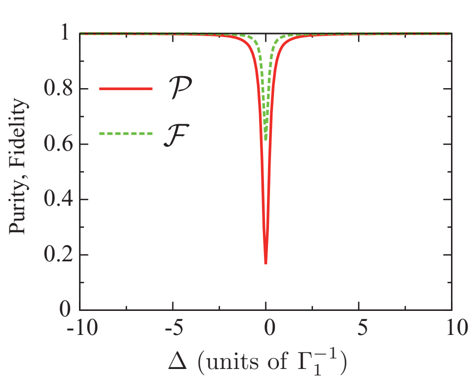

FIG. 8. (Color online) $\mathcal{P}$ and $\mathcal{F}$ as functions of the detuning Δ. The pulse length is fixed at $d = 100\Gamma_1^{-1}$.

ACKNOWLEDGMENTS

The authors are grateful to Professor T. Kato for fruitful discussions. This research was partially supported by the Nakajima Foundation and MEXT KAKENHI (17GS1204, 21104507).

APPENDIX: SOLUTION OF EQUATION OF MOTION

To solve the Schrödinger equation, we use the coherent state $|\phi_{\text{in}}\rangle$ as the input state. Since we consider the input state, which is composed of the horizontally polarized photons, the state vector of a coherent state is given by

$$|\phi_{\text{in}}\rangle = \mathrm{e}^{-|\mu|^2/2}\exp\left[\mu \int dr f(r)\tilde{h}_r^\dagger\right]|0\rangle. \tag{A1}$$

The output state vector for $|\phi_{\text{in}}\rangle$ is obtained by the Schrödinger equation $|\phi_{\text{out}}\rangle = e^{-i\mathcal{H}t}|\phi_{\text{in}}\rangle$. Expanding in powers of μ, the state vector $|\phi_{\text{in(out)}}\rangle$ can be rewritten as

$$|\phi_{\text{in(out)}}\rangle = |0\rangle + \mu\left|\psi_{\text{in(out)}}^{(1)}\right\rangle + \frac{\mu^2}{\sqrt{2}}\left|\psi_{\text{in(out)}}^{(2)}\right\rangle + \cdots. \tag{A2}$$

The linear and quadratic components of $|\phi_{\text{in(out)}}\rangle$ give $|\psi_{\text{in(out)}}^{(1)}\rangle$ and $|\psi_{\text{in(out)}}^{(2)}\rangle$, respectively. Using the coherent output state, we can obtain the one- and two-photon output wave functions for single-photon input from

$$\langle\tilde{h}_r\rangle^{(1)} = g_{1\to1}(r;t), \tag{A3}$$

$$\left\langle\tilde{v}_{r_1}\tilde{v}_{r_2}\right\rangle^{(1)} = \sqrt{2}g_{1\to2}(r_1,r_2;t), \tag{A4}$$

where we define the average $\langle A\rangle^{(1)}$ as the linear component of $\langle\phi_{\text{out}}|A|\phi_{\text{out}}\rangle$. Likewise, the three-photon output wave function for two input photons is obtained by

$$\left\langle\tilde{h}_r\tilde{v}_{r_1}\tilde{v}_{r_2}\right\rangle^{(2)} = \sqrt{2}g_{2\to3}(r,r_1,r_2;t), \tag{A5}$$

where the average $\langle A\rangle^{(2)}$ represents the quadratic component of $\langle\phi_{\text{out}}|A|\phi_{\text{out}}\rangle$.

From the Heisenberg equations for $\tilde{h}_k(t)$ and $\tilde{v}_k(t)$, the output field operators in the range of $0 < r < t$ are given by

$$\tilde{h}_r(t) = \tilde{h}_{-t-r}(0) - \sqrt{\Gamma_1}\sigma_{ge}(t-r), \tag{A6}$$

$$\tilde{v}_r(t) = \tilde{v}_{-t}(0) - \sqrt{\Gamma_2}\sigma_{me}(t-r) - \sqrt{\Gamma_3}\sigma_{gm}(t-r), \tag{A7}$$

where $\tilde{h}_{-t}(0)$ and $\tilde{v}_{-t}(0)$ are initial field operators at the position $r = -t$. Using the above input-output relations, the output wave functions for one input photon, which are given by Eqs. (A3) and (A4), can be rewritten as

$$g_{1\to1}(r;t) = f(-t-r) - \sqrt{\Gamma_1}\langle\sigma_{ge}(t-r)\rangle^{(1)}, \quad \text{(A8)}$$

$$g_{1\to2}(r_1,r_2;t) = \sqrt{\frac{\Gamma_2\Gamma_3}{2}}[\langle\sigma_{me}(t-r_1)\sigma_{gm}(t-r_2)\rangle^{(1)} + \langle\sigma_{gm}(t-r_1)\sigma_{me}(t-r_2)\rangle^{(1)}]. \quad \text{(A9)}$$

Here, we use the relation $\tilde{h}_r(0)|\phi_{\rm in}\rangle = \mu f(r)|\phi_{\rm in}\rangle$ and $\tilde{v}_r(0)|\phi_{\rm in}\rangle = 0$. In order to calculate $\langle\sigma_{ge}(t-r)\rangle^{(1)}$, $\langle\sigma_{me}(t-r_1)\sigma_{gm}(t-r_2)\rangle^{(1)}$, and $\langle\sigma_{gm}(t-r_1)\sigma_{me}(t-r_2)\rangle^{(1)}$, we work in the Heisenberg picture. The Heisenberg equations for σ_{gm} and σ_{ge} are written as

$$\frac{d}{dt}\sigma_{gm} = \left[-i\Omega_m - \frac{\Gamma_3}{2}\right]\sigma_{gm} + \sqrt{\Gamma_2\Gamma_3}\sigma_{me} - \sqrt{\Gamma_1}\sigma_{em}\tilde{h}_{-t}(0) - \sqrt{\Gamma_2}\tilde{v}^{\dagger}_{-t}(0)\sigma_{ge} + \sqrt{\Gamma_3}(\sigma_{gg}-\sigma_{mm})\tilde{v}_{-t}(0), \quad \text{(A10)}$$

$$\frac{d}{dt}\sigma_{ge} = \left[-i\Omega_e - \frac{\Gamma_1+\Gamma_2}{2}\right]\sigma_{ge} + \sqrt{\Gamma_1}(\sigma_{gg}-\sigma_{ee})\tilde{h}_{-t}(0) + \sqrt{\Gamma_2}\sigma_{gm}\tilde{v}_{-t}(0) - \sqrt{\Gamma_3}\sigma_{me}\tilde{v}_{-t}(0). \quad \text{(A11)}$$

Using the initial condition where only $\langle\sigma_{gg}\rangle$ has a finite value in the zeroth order for μ, we can solve the equations of motion for $\langle\sigma_{ge}(t)\rangle^{(1)}$, $\langle\sigma_{gm}(t-r_1)\sigma_{me}(t-r_2)\rangle^{(1)}$, and $\langle\sigma_{me}(t-r_1)\sigma_{gm}(t-r_2)\rangle^{(1)}$, respectively:

$$\langle\sigma_{ge}(t)\rangle^{(1)} = \sqrt{\Gamma_1}\int_0^{\infty} d\xi f(-t+\xi)e^{-i\tilde{\Omega}_e\xi}, \quad \text{(A12)}$$

$$\langle\sigma_{gm}(t-r_1)\sigma_{me}(t-r_2)\rangle^{(1)} = \langle\sigma_{ge}(t-r_2)\rangle^{(1)}e^{-i\tilde{\Omega}_m(r_2-r_1)}, \quad \text{(A13)}$$

$$\langle\sigma_{me}(t-r_1)\sigma_{gm}(t-r_2)\rangle^{(1)} = 0, \quad \text{(A14)}$$

where the frequencies $\tilde{\Omega}_e$ and $\tilde{\Omega}_m$ denote $\Omega_e - i(\Gamma_1+\Gamma_2)/2$ and $\Omega_m - i\Gamma_3/2$, respectively, and we set $r_1 \leqslant r_2$. Substituting Eqs. (A12)–(A14) into Eqs. (A8) and (A9), we obtain one- and two-photon output wave functions for a single input photon ($g_{1\to1}$ and $g_{1\to2}$).

The two-, three-, and four-photon output wave functions for two input photons are given by

$$g_{2\to2}(r_1,r_2;t) = g_{1\to1}(r_1;t)g_{1\to1}(r_2;t) + \delta g_{2\to2}(r_1,r_2;t), \quad \text{(A15)}$$

$$g_{2\to3}(r,r_1,r_2;t) = g_{1\to1}(r;t)g_{1\to2}(r_1,r_2;t) + \delta g_{2\to3}(r,r_1,r_2;t), \quad \text{(A16)}$$

$$g_{2\to4}(r_1,r_2,r_3,r_4;t) = g_{1\to2}(r_1,r_2;t)g_{1\to2}(r_3,r_4;t) + \delta g_{2\to4}(r_1,r_2,r_3,r_4;t). \quad \text{(A17)}$$

Equations (A5)–(A7) show that $\delta g_{2\to2}$, $\delta g_{2\to3}$, and $\delta g_{2\to4}$ contain many terms. However, by solving the Heisenberg equations, we can straightforwardly obtain the following equations for a quadratic order of μ:

$$\delta g_{2\to2}(r_1,r_2;t) = -\Gamma_1[\langle\sigma_{ge}(t-r_2)\rangle^{(1)}]^2 e^{-i\tilde{\Omega}_e(r_2-r_1)}, \quad \text{(A18)}$$

$$\delta g_{2\to3}(r,r_1,r_2;t) = \begin{cases} \sqrt{\Gamma_1\Gamma_2\Gamma_3}\langle\sigma_{ge}(t-r_1)\rangle^{(1)}\langle\sigma_{gm}(t-r_1)\sigma_{me}(t-r_2)\rangle^{(1)}e^{-i\tilde{\Omega}_e(r_1-r)} & (r\leqslant r_1\leqslant r_2)\\ \sqrt{\Gamma_1\Gamma_2\Gamma_3}\langle\sigma_{ge}(t-r)\rangle^{(1)}\langle\sigma_{gm}(t-r)\sigma_{me}(t-r_2)\rangle^{(1)}e^{-i\tilde{\Omega}_m(r-r_1)} & (r_1\leqslant r\leqslant r_2)\\ \sqrt{\Gamma_1\Gamma_2\Gamma_3}\langle\sigma_{ge}(t-r)\rangle^{(1)}\langle\sigma_{gm}(t-r_2)\sigma_{me}(t-r_1)\rangle^{(1)}e^{-i\tilde{\Omega}_e(r-r_2)} & (r_1\leqslant r_2\leqslant r), \end{cases} \quad \text{(A19)}$$

$$\delta g_{2\to4}(r_1,r_2,r_3,r_4;t) = -\left[g^2_{1\to2}(r_3,r_4;t)\right]e^{-i\tilde{\Omega}_m(r_2-r_1)}e^{-i\tilde{\Omega}_e(r_3-r_2)}e^{-i\tilde{\Omega}_m(r_4-r_3)}. \quad \text{(A20)}$$

Thus, we obtain the two-, three- and four-photon output wave functions by substituting Eqs. (A18)–(A20) into Eqs. (A15)–(A17), respectively.

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