Two-photon nonlinearity in general cavity QED systems

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We have investigated two-photon nonlinearity in general cavity QED systems, which cover both weak- and strong-coupling regimes and include the radiative loss from an atom. One- and two-photon propagators are obtained in analytical forms. By surveying both coupling regimes, we have revealed the conditions on a photonic pulse for yielding large nonlinear effects, depending on the cavity Q value. We also discuss the effects of radiative loss on nonlinearity.

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I. INTRODUCTION

Significant nonlinear optical effects are usually obtained when intense light fields are irradiated onto a material with large nonlinear susceptibilities. By using an optical cavity, it is possible to magnify the electric field inside a cavity, provided that the input field is resonant with the cavity mode [1]. Thus, it is possible to enhance the nonlinear optical response by placing the nonlinear material inside a cavity. This idea was realized experimentally by using two-level atoms as the nonlinear material [2,3], which demonstrated the possibility of obtaining large nonlinearity even using weak input fields. In particular, nonlinear effects appearing in the two-photon state have recently attracted much attention due to possible applications for quantum information devices [4] and also due to recent progress in the photon-pair manipulation technique [5,6].

In order to discuss theoretically the dynamics of the two-photon state, a quantum-mechanical treatment of the photon field is indispensable, because the nonlinearity appears in the wave function of the photon, not in the amplitude [7]. Such an analysis was pioneered in a case in which the nonlinear material is a one-dimensional atom, obtained as the lossless and weak-coupling limit of an atom-cavity system [8]. However, considering that magnification of photon fields occurs more effectively in better (higher-Q-value) cavities, strong-coupling cases also seem promising for yielding large nonlinearity [9]. The optimum cavity conditions for two-photon nonlinearity have not been sufficiently discussed in the literature; thus, this type of nonlinearity should be investigated using methods applicable to any coupling regime. In the present study, we present analytical expressions for one- and two-photon propagators in general atom-cavity systems, in which both the weak- and strong-coupling regimes are covered and the loss from the cavity due to radiative decay into noncavity modes is taken into account. Our discussion of two-photon nonlinearity is based on the use of this propagator. Moreover, we clarify the optimum conditions on photonic pulses for achieving large nonlinearity, depending on the cavity conditions.

This study is presented as follows. In Sec. II, the theoretical model of the atom-cavity system is introduced, and the significance of the parameters is described. In Sec. III, we define the input and output states of the photons. In Sec. IV, the measure of the nonlinearity appearing in the output state is defined. In Sec. V, we describe the form of the input wave function and a scaling law for this atom-cavity system. In Sec. VI, we numerically evaluate the nonlinearity for lossless cases and clarify the optimum conditions for inducing large nonlinearity. In Sec. VII, the effects of loss are discussed. The analytic expressions for the one- and two-photon propagators are shown in the Appendix.

II. SYSTEM

In this study, we investigate a single two-level system (hereafter referred to as an "atom") embedded in a one-sided cavity [10] as a nonlinear optical system. The system is schematically illustrated in Fig. 1. The atom is coupled not only to the cavity mode but also to the noncavity modes. The cavity mode is coupled through the right mirror of the cavity to the external photon field, which is labeled one-dimensionally by r. Although the external field actually extends only into the $r>0$ region and the incoming and outgo-

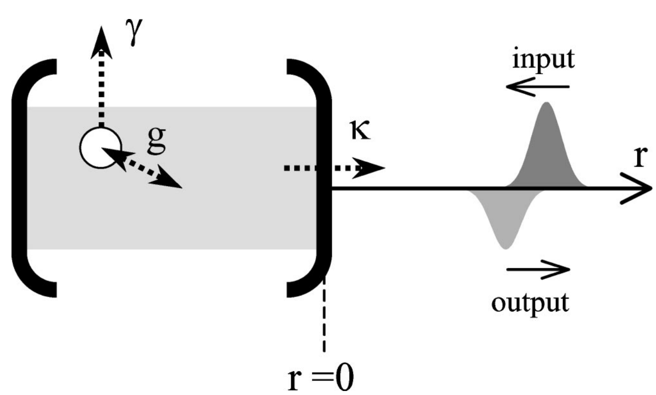

FIG. 1. Schematic view of the atom-cavity system. The right mirror of the cavity is weakly transmissive, through which the cavity mode is coupled to the external photon field. g,γ,κ represent the atom-cavity coupling, the radiative decay rate of the atom into noncavity modes, and the decay rate of the cavity mode, respectively.

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ing photons are traveling in the opposite direction, we may treat the incoming photons as if they were propagating in the $r<0$ region in the positive direction [11].

The Hamiltonian of the system is given, setting $\hbar=c=1$, by

$$\mathcal{H} = \omega_a \sigma_+ \sigma_- + \int d\mu \mu d_\mu^\dagger d_\mu + \int d\mu (\gamma_\mu \sigma_+ d_\mu + \text{H.c.}) + \omega_c c^\dagger c + \int dk k b_k^\dagger b_k + \int dk (\kappa_k c^\dagger b_k + \text{H.c.}) + g(\sigma_+ c + c^\dagger \sigma_-), \tag{1}$$

where σ_-, c, b_k, and d_μ are the annihilation operators for the atomic excitation, cavity mode, external photon mode, and noncavity mode, respectively. ω_a and ω_c represent the frequencies of the atomic transition and the cavity mode, and g represents the atom-cavity coupling. Regarding γ_μ and κ_k, we use the flatband assumption, i.e., $\gamma_\mu=(\gamma/2\pi)^{1/2}$ and $\kappa_k=(\kappa/2\pi)^{1/2}$, by which the damping rates of the atom and the cavity mode are given by γ and κ.

The complex frequencies of the atom and cavity are defined by the following equations:

$$\tilde{\omega}_a = \omega_a - i\gamma/2, \tag{2}$$

$$\tilde{\omega}_c = \omega_c - i\kappa/2. \tag{3}$$

Using these frequencies, the complex eigenfrequencies $\tilde{\omega}_{1,2}$ of the atom-cavity system are given by

$$(\omega - \tilde{\omega}_1)(\omega - \tilde{\omega}_2) = (\omega - \tilde{\omega}_a)(\omega - \tilde{\omega}_c) - g^2. \tag{4}$$

It is of note that γ is related to the dissipation of this atom-cavity system. When $\gamma=0$, the input photons are always reflected back into the output port, i.e., the atom-cavity system is lossless. In contrast, when $\gamma\neq 0$, some of the input photons are lost as spontaneous emission into the noncavity modes, i.e., the atom-cavity system is lossy. In the limit of $\gamma\to 0$ and $(g,\kappa)\to\infty$, keeping $\Gamma=4g^2/\kappa$ constant, this atom-cavity system is reduced to a one-dimensional atom, where the atom is directly coupled to the external field at $r=0$ with the coupling constant Γ [2,8].

The real-space operator of the external field, b_r, is given by the Fourier transform of b_k:

$$b_r = (2\pi)^{-1/2} \int dk e^{ikr} b_k. \tag{5}$$

We note again that the negative (positive) r represents the incoming (outgoing) field.

III. INPUT AND OUTPUT STATES

Our main concern in this study is to determine how the initial one- or two-photon wave functions are transformed after interacting with the atom-cavity system. In the initial state, the atom and the cavity are in the ground states and one or two photons exist in the input port, i.e.,

$$|\Psi_{\text{in}}^{(1)}\rangle = \int dr \psi_{\text{in}}(r) b_r^\dagger |0\rangle, \tag{6}$$

$$|\Psi_{\text{in}}^{(2)}\rangle = 2^{-1/2} \int dr_1 dr_2 \psi_{\text{in}}(r_1,r_2) b_{r_1}^\dagger b_{r_2}^\dagger |0\rangle. \tag{7}$$

The one-photon wave function $\psi_{\text{in}}(r)$ is normalized as $\int dr|\psi_{\text{in}}(r)|^2=1$, and $\psi_{\text{in}}(r)=0$ in $r>0$. Similarly, $\psi_{\text{in}}(r_1,r_2)$ satisfies $\int dr_1 dr_2 |\psi_{\text{in}}(r_1,r_2)|^2=1$ and $\psi_{\text{in}}(r_1,r_2)=0$ in $r_1,r_2>0$, and has the following symmetry: $\psi_{\text{in}}(r_2,r_1)=\psi_{\text{in}}(r_1,r_2)$.

After the photons have interacted with this atom-cavity system, the excitations in the atom and the cavity mode completely escape to the external modes (b_k) or to the noncavity modes (d_μ). Then the states of the system are written as follows:

$$|\Psi_{\text{out}}^{(1)}\rangle = \int dr \psi_{\text{out}}(r) b_r^\dagger |0\rangle + \cdots, \tag{8}$$

$$|\Psi_{\text{out}}^{(2)}\rangle = 2^{-1/2} \int dr_1 dr_2 \psi_{\text{out}}(r_1,r_2) b_{r_1}^\dagger b_{r_2}^\dagger |0\rangle + \cdots, \tag{9}$$

where the ellipses imply the terms containing excitations in the noncavity modes. Whereas the number of photons is conserved in the output pulse $[\int dr|\psi_{\text{out}}(r)|^2=1$ and $\int dr_1 dr_2|\psi_{\text{out}}(r_1,r_2)|^2=1]$ in the case of $\gamma=0$, the output state is attenuated in general cases $(\gamma\neq 0)$.

The input and output wave functions are related by the propagator G as follows:

$$\psi_{\text{out}}(r) = \int dr' G(r;r') \psi_{\text{in}}(r'), \tag{10}$$

$$\psi_{\text{out}}(r_1,r_2) = \int dr_1' dr_2' G(r_1,r_2;r_1',r_2') \psi_{\text{in}}(r_1',r_2'). \tag{11}$$

The one- and two-photon propagators are analytically obtainable for this atom-cavity system (see the Appendix). The two-photon propagator has the following symmetry: $G(r_2,r_1;r_2',r_1')=G(r_1,r_2;r_1',r_2')$, which guarantees the symmetry in the output wave function, $\psi_{\text{out}}(r_2,r_1)=\psi_{\text{out}}(r_1,r_2)$.

IV. MEASURE OF NONLINEARITY

When two photons are input into this atom-cavity system, the input wave function $\psi_{\text{in}}(r_1,r_2)$ is finally transformed to the output wave function $\psi_{\text{out}}(r_1,r_2)$. In order to quantify the nonlinear effect in this process, we compare $\psi_{\text{out}}(r_1,r_2)$ with the *linear* output wave function $\psi_{\text{out}}^{\text{L}}(r_1,r_2)$, which is defined by

$$\psi_{\text{out}}^{\text{L}}(r_1,r_2) = \int dr_1' dr_2' G(r_1;r_1') G(r_2;r_2') \psi_{\text{in}}(r_1',r_2'), \tag{12}$$

where $G(r;r')$ is the one-photon propagator. This linear output is obtained when the atom in the cavity is replaced by a harmonic oscillator with the same transition frequency, i.e., when the nonlinearity of the system is completely removed.

We here define the quantity β as follows:

$$\beta = \frac{\int dr_1 dr_2 (\psi_{\mathrm{out}}^{\mathrm{L}})^* \psi_{\mathrm{out}}}{\sqrt{\int dr_1 dr_2 |\psi_{\mathrm{out}}|^2}\sqrt{\int dr_1 dr_2 |\psi_{\mathrm{out}}^{\mathrm{L}}|^2}}, \qquad (13)$$

which gives the *angle* between ψ_{out} and $\psi_{\mathrm{out}}^{\mathrm{L}}$. β always lies in the unit circle ($|\beta| \leq 1$) due to Schwarz's inequality, and $\beta = 1$ holds when the response of the system is completely linear ($\psi_{\mathrm{out}} = \psi_{\mathrm{out}}^{\mathrm{L}}$). Thus, the nonlinear effect is reflected in the deviation of β from unity, and $|\beta - 1|$ may be regarded as a measure of the nonlinear effect. In addition, when the atom-cavity system is lossless, $\int dr_1 dr_2 |\psi_{\mathrm{out}}|^2 = \int dr_1 dr_2 |\psi_{\mathrm{out}}^{\mathrm{L}}|^2 = 1$ and β is simply reduced to the overlap integral between ψ_{out} and $\psi_{\mathrm{out}}^{\mathrm{L}}$.

V. INPUT WAVE FUNCTIONS AND SCALING LAW

Now we employ the above formalism for specific forms of the input states and clarify the conditions required to obtain a large degree of nonlinearity. In this study, we focus on a case in which the two input photons have the same wave function of Gaussian form, i.e.,

$$\psi_{\mathrm{in}}(r_1, r_2) = \psi_{\mathrm{in}}(r_1 - a)\psi_{\mathrm{in}}(r_2 - a), \qquad (14)$$

$$\psi_{\mathrm{in}}(r) = (2/\pi d^2)^{1/4} \exp(-r^2/d^2 + iqr). \qquad (15)$$

The input wave function is characterized by two parameters, q (the central frequency) and d (the coherent length of the photon). $a(<0)$ in Eq. (14) represents the initial position of the photons, which is an irrelevant parameter.

The atom-cavity system is characterized by g (the coupling between the atom and the cavity mode), $\omega_a - \omega_c$ (the frequency difference between the atom and the cavity mode), γ (the damping rate of the atom into the noncavity modes), and κ (the damping rate of the cavity mode). Adding the input-photon parameters q and d, the whole system is specified by the set of parameters $(g, \omega_a - \omega_c, \gamma, \kappa, q, d)$.

The number of parameters may be decreased with the help of the following scaling law. Consider a scaling transformation $\mathcal{H} \rightarrow \alpha\mathcal{H}$, where α is a positive constant. The scaled Hamiltonian takes the form

$$\alpha\mathcal{H} = \alpha\omega_a\sigma_+\sigma_- + \int d\mu \mu D_\mu^\dagger D_\mu + \int d\mu(\alpha^{1/2}\gamma_\mu \sigma_+ D_\mu + \mathrm{H.c.}) + \alpha\omega_c c^\dagger c + \int dk k B_k^\dagger B_k + \int dk(\alpha^{1/2}\kappa_k c^\dagger B_k + \mathrm{H.c.}) + \alpha g(\sigma_+ c + c^\dagger \sigma_-), \qquad (16)$$

where $B_k = \alpha^{-1/2} b_{k/\alpha}$ and $D_\mu = \alpha^{-1/2} d_{\mu/\alpha}$ satisfy the orthonormalized commutation relations $[B_k, B_{k'}^\dagger] = \delta(k - k')$, etc. As for the photonic wave functions, noticing that $b_r = \alpha^{-1/2} B_{r/\alpha}$, they are transformed as $\psi(r) \rightarrow \alpha^{1/2}\psi(\alpha r)$. Comparing the original and scaled quantities, it is concluded that a system with parameters $(\alpha g, \alpha(\omega_a - \omega_c), \alpha\gamma, \alpha\kappa, \alpha q, d/\alpha)$ is equivalent to a system with parameters $(g, \omega_a - \omega_c, \gamma, \kappa, q, d)$. Using this law, the system is specified by the following set of dimensionless parameters: $((\omega_a - \omega_c)/g, \gamma/g, \kappa/g, q/g, gd)$. In the following numerical examples, $\omega_a - \omega_c$ is fixed at zero and ω_a ($=\omega_c$) is chosen as the origin of frequency.

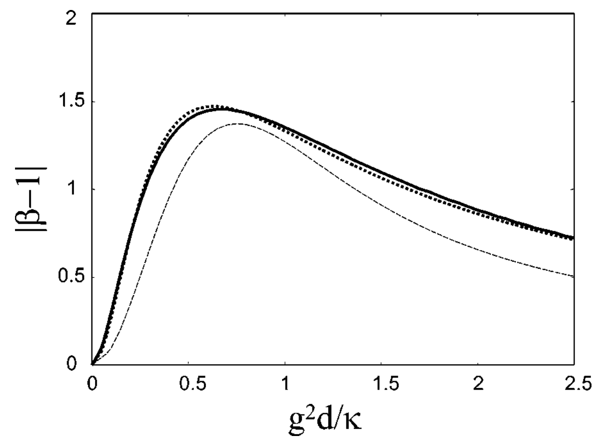

FIG. 2. Dependence of $|\beta - 1|$ on $g^2 d/\kappa$ in the weak-coupling regime [$\kappa/g = 10$ (solid line) and $\kappa/g = 5$ (dotted line)]. The frequency of the photons is chosen at $q = 0$. In the strong-coupling regime, the $q = 0$ photons are no longer resonant due to the Rabi splitting and the nonlinearity becomes smaller (thin broken line, where $\kappa/g = 2$).

VI. NUMERICAL RESULTS FOR LOSSLESS CASES

In this section, we present the numerical results obtained for the $\gamma = 0$ cases, in which the atom-cavity system is lossless and the number of photons is preserved in the output pulse. The atom-cavity system is then characterized solely by the ratio κ/g, as was discussed in Sec. V. Equation (4) indicates that Rabi splitting of the eigenfrequency of the atom-cavity system takes place when $\kappa/g < 4$. Thus, in our definition of the parameters, the strong- (weak-)coupling regime is specified by the inequality $\kappa/g \lesssim 4$ ($\kappa/g \gtrsim 4$).

A. Weak-coupling regime

First, we discuss the weak-coupling regime. In Fig. 2, the nonlinearity $|\beta - 1|$ is plotted for cases of resonant input ($q = 0$) as a function of κ/g and $g^2 d/\kappa$. It has been confirmed that, in the weak-coupling regime, nonlinearity appears most strongly at $q = 0$ and gets weaker as $|q|$ is increased.

Figure 2 indicates that, roughly speaking, $|\beta - 1|$ is dependent solely on $g^2 d/\kappa$ in the weak-coupling regime. This fact can be explained as follows. In the weak-coupling regime, the atom-cavity system may be regarded as a "one-dimensional atom," where the atom is coupled directly to a one-dimensional photon field with a coupling constant of $4g^2/\kappa$. The figure also clarifies that the nonlinearity is maximized when $g^2 d/\kappa \sim 0.5$. For example, for $g = 120$ MHz and $\kappa = 900$ MHz [3], the optimum pulse length d is about 9 m. The qualitative explanation for this optimum condition will be given in Sec. VI C.

B. Strong-coupling regime

Next, we discuss the strong-coupling regime. In Fig. 3, the nonlinearity $|\beta - 1|$ is plotted for fixed κ/g($=0.5$) as a

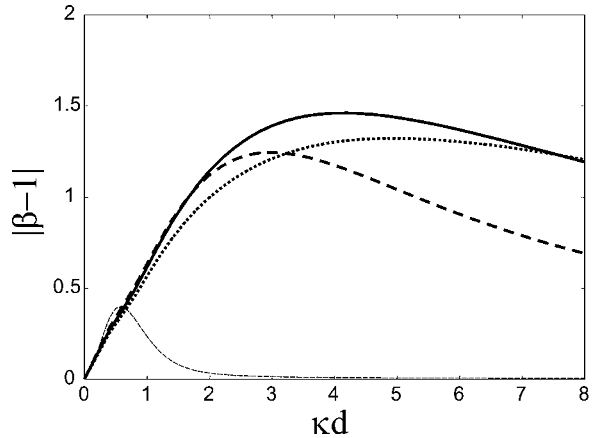

FIG. 3. Dependence of $|\beta-1|$ on κd, where κ/g is fixed at 0.5 (strong-coupling regime). $q/g=0.8$ (broken line), $q/g=0.9$ (solid line), $q/g=1$ (dotted line), and $q/g=0$ (thin broken line).

function of κd and q/g. In contrast to the weak-coupling regime, the nonlinearity is weak for resonant input ($q=0$). This is because the $q=0$ photons are no longer resonant with the cavity mode due to the Rabi splitting. Instead, a large nonlinearity is obtained when the input photons are tuned to the Rabi-split frequency, $q\sim\pm g$. The maximum value of $|\beta-1|$ is approximately 1.5, which is almost the same as the value obtained in the weak-coupling regime.The nonlinearity is optimized for $\kappa d\sim 4$, the reasons for which will be discussed in Sec. VI C.

C. Optimum pulse for inducing large nonlinearity

In the preceding subsections, we clarified the optimum frequency q and the length d of the photon pulse required for inducing large nonlinearity in both weak- and strong-coupling regimes. Here, we account for these optimum conditions from a unified perspective. To this end, we focus on the wave function $\varphi(r)$, which is defined by

$$e^{i\mathcal{H}t}\sigma_+|0\rangle = \int dr\varphi(r)b_r^\dagger|0\rangle \qquad (17)$$

for a large $t(>0)$. The significance of $\varphi(r)$ becomes clear by multiplying $e^{-i\mathcal{H}t}$ from the left; if a single photon pulse $\varphi(r)$ is prepared as an input at time t_0, the photon will be completely absorbed by the atom at time t_0+t. Therefore, when the input pulse $\psi_{\rm in}(r)$ resembles $\varphi(r)$ in shape, the two input photons try to occupy the atom simultaneously and strong nonlinear effects are expected.

$\varphi(r)$ has the following form:

$$\varphi(r) = \begin{cases} \dfrac{ig\kappa^{1/2}}{\tilde{\omega}_1^*-\tilde{\omega}_2^*}(e^{i\tilde{\omega}_1^*(r+t)}-e^{i\tilde{\omega}_2^*(r+t)}) \quad (-t<r<0), \\ 0 \quad (\text{otherwise}), \end{cases} \qquad (18)$$

where $\tilde{\omega}_1$ and $\tilde{\omega}_2$ are the complex eigenfrequencies of the atom-cavity system, as defined in Eq. (4). In the weak-coupling regime, $\tilde{\omega}_1$ and $\tilde{\omega}_2$ are approximately given by

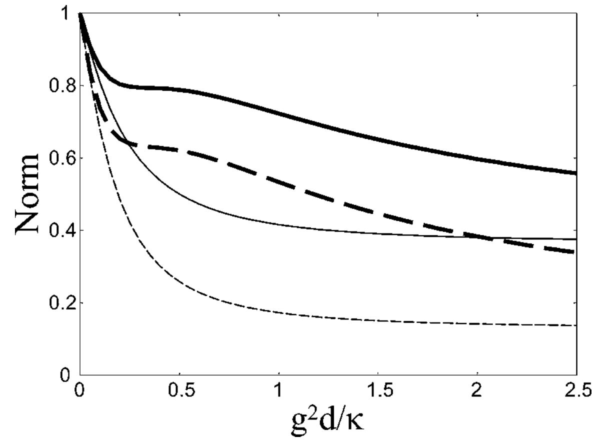

FIG. 4. Norms of $\psi_{\rm out}(r_1,r_2)$ (bold lines) and $\psi^{\rm L}_{\rm out}(r_1,r_2)$ (thin lines), for lossy cases of $\gamma/g=0.1$ (solid lines) and $\gamma/g=0.2$ (broken lines). The atom-cavity system is in the weak-coupling regime ($\kappa/g=5$) and resonant photons ($q=0$) are used.

$-i\kappa/2$ and $-2ig^2/\kappa$. Because $g^2/\kappa\gg\kappa$ in this regime, the optimum frequency q and pulse length d are given by $q\sim 0$ and $d\sim\kappa/2g^2$, which accounts for the numerical results in Sec. VI A. On the other hand, in the strong-coupling regime, $\tilde{\omega}_1$ and $\tilde{\omega}_2$ are approximately given by $-i\kappa/4\pm g$. Therefore, the optimum q and d are roughly estimated at $q\sim\pm g$ and $d\sim 4/\kappa$, which is in agreement with the numerical results given in Sec. VI B.

VII. NUMERICAL RESULTS FOR LOSSY CASES

In the preceding section, the results obtained for the lossless cases ($\gamma=0$) are presented. Here, we discuss the lossy cases ($\gamma\neq 0$), fixing the parameters $\kappa/g=5$ (weak-coupling regime) and $q=0$, which serve as an example.

The apparent result of the loss is attenuation of the photon pulses. In Fig. 4, we have plotted the norm of the two-photon output wave function $\psi_{\rm out}(r_1,r_2)$, i.e., the probability of finding two photons in the output. The norm of the linear output wave function $\psi^{\rm L}_{\rm out}(r_1,r_2)$ is also plotted in the same figure. As expected, the photon pulse is attenuated more significantly when the loss parameter γ is larger, for both $\psi_{\rm out}$ and $\psi^{\rm L}_{\rm out}$ (compare the solid and broken lines). It is also observed that $\psi^{\rm L}_{\rm out}$ is more attenuated than $\psi_{\rm out}$. This tendency can be understood by the facts that, in the linear case, the photons are more likely to be absorbed by the atom due to the absence of the saturation effect, and that the loss of photons occurs only while the photons are occupying the atom, as schematically shown in Fig. 1.

Figure 5 plots the nonlinear parameter $|\beta-1|$ defined in Eq. (13), when the system is lossy, i.e., $\gamma\neq 0$. It is observed that $|\beta-1|$ decreases slightly for the $g^2d/\kappa\lesssim 0.5$ region and increases slightly for the $g^2d/\kappa\gtrsim 0.5$ region; however, in general, no qualitative changes are introduced by the loss. Thus, although the probability of finding two photons in the output decreases significantly as shown in Fig. 4, the nonlinear characteristics of the output state are almost unchanged if the output pulse contains two photons.

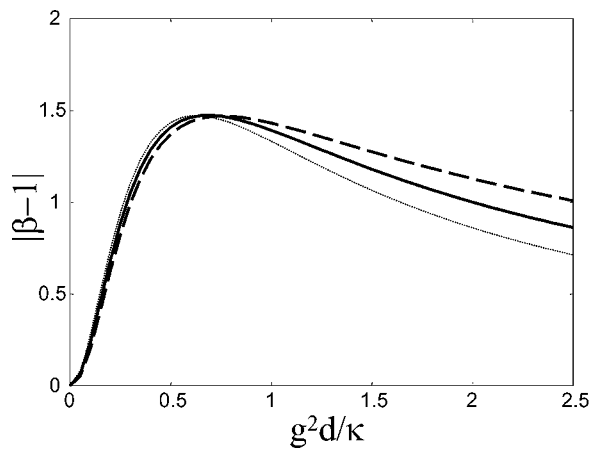

FIG. 5. The effect of loss on the nonlinearity $|\beta-1|$; $\gamma/g=0$ (thin dotted line), $\gamma/g=0.1$ (solid line), and $\gamma/g=0.2$ (broken line). The other parameters are $\kappa/g=5$ and $q=0$.

VIII. SUMMARY

Here, we theoretically investigated a situation in which two photons were simultaneously input into a nonlinear optical system, and we examined the nonlinear effect in the output wave function. As a model nonlinear optical system, we employed a two-level system (atom) embedded in a cavity, which is illustrated in Fig. 1. The one- and two-photon propagators were obtained in the analytic forms that are presented in the Appendix. The angle between the linear and nonlinear two-photon output wave functions, given by Eq. (13), was used to measure the nonlinear effect. This quantity was numerically evaluated in the weak-coupling regime (Fig. 2) and in the strong-coupling regime (Fig. 3), and the conditions for inducing large nonlinearity were revealed. These conditions were explained by the optimum pulse shape, given by Eq. (18). The present findings suggest that pulse shape control is more essential for optimizing the two-photon nonlinearity than Q-value control of the cavity system. We also considered $\gamma\neq 0$ cases, in which the atom-cavity system is lossy. As shown in Fig. 4, the probability of finding two photons in the output decreased greatly due to the loss effect, but the nonlinearity in the output two-photon wave function remained almost unchanged in comparison with the lossless case.

ACKNOWLEDGMENTS

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APPENDIX: PROPAGATORS

In this appendix, we present the analytic forms of the one- and two-photon propagators $G(r;r')$ and $G(r_1,r_2;r_1',r_2')$. Using the completeness relation in the one-photon space, $\hat{1}=\int dr b_r^\dagger|0\rangle\langle 0|b_r$, the one-photon propagator is given by

$$G(r;r')=\langle 0|b_r e^{-i\mathcal{H}t}b_{r'}^\dagger|0\rangle, \tag{A1}$$

where $r>0>r'$, and t is the time difference between the output and the input. This propagator is derivable by standard application of the Green function method [12]. Throughout this section, we employ a coordinate system moving at light speed; transformation to a static coordinate system is carried out by replacing r (coordinates without primes) with $r-t$, or by replacing r' (coordinates with primes) with $r'+t$. In this coordinate system, the one-photon propagator is given as follows:

$$G(r,r')=G(r-r')=G_0(r-r')+G_2(r-r'), \tag{A2}$$

$$G_0(r-r')=\delta(r-r')-\kappa\theta(r'-r)e^{i\tilde{\omega}_c(r-r')}, \tag{A3}$$

$$G_2(r-r')=\kappa\theta(r'-r)\left(e^{i\tilde{\omega}_c(r-r')}-\frac{\tilde{\omega}_2-\tilde{\omega}_c}{\tilde{\omega}_2-\tilde{\omega}_1}e^{i\tilde{\omega}_1(r-r')}-\frac{\tilde{\omega}_1-\tilde{\omega}_c}{\tilde{\omega}_1-\tilde{\omega}_2}e^{i\tilde{\omega}_2(r-r')}\right), \tag{A4}$$

where $\tilde{\omega}_a$ (complex frequency of the atom), $\tilde{\omega}_c$ (complex frequency of the cavity), and $\tilde{\omega}_{1,2}$ (complex eigenfrequencies of the atom-cavity system) are defined in Eqs. (2)–(4), respectively.

Next, we proceed to the two-photon case. Using the completeness relation in the two-photon space, $\hat{1}=2^{-1}\int dr_1 dr_2 b_{r_1}^\dagger b_{r_2}^\dagger|0\rangle\langle 0|b_{r_1}b_{r_2}$, the two-photon propagator is identified as $2^{-1}\langle 0|b_{r_1}b_{r_2}e^{-i\mathcal{H}t}b_{r_1'}^\dagger b_{r_2'}^\dagger|0\rangle$. This quantity is composed of two types of terms; in the first (second) type, the photons initially located at r_1' and r_2' are scattered to r_1 and r_2 (r_2 and r_1), respectively. With the help of $\psi_{\rm in}(r_1',r_2')=\psi_{\rm in}(r_2',r_1')$, it can be shown that both types of terms yield the same output wave function $\psi_{\rm out}(r_1,r_2)$ after integration in Eq. (11). We can therefore safely regard only the first type of terms as constituting the two-photon propagator. The two-photon propagator is thus given by

$$G(r_1,r_2,r_1',r_2')=G(r_1-r_1')G(r_2-r_2')-G_2(r_1-r_1')G_2(r_2-r_2')+\sum_{j=4,6,8}G_j(r_1,r_2,r_1',r_2'), \tag{A5}$$

where G_j $(j=4,6,8)$ are defined by

$$G_4(r_1,r_2,r_1',r_2') = \frac{-ig^4\kappa^2}{8\pi^3}[I_4(r_1-r_2,r_1'-r_2',r_2-r_1') + I_4(r_2-r_1,r_2'-r_1',r_1-r_2')], \tag{A6}$$

$$G_6(r_1,r_2,r_1',r_2') = \frac{-ig^6\kappa^2}{8\pi^3}[I_6(r_2-r_1,r_1'-r_2',r_1-r_1') + I_6(r_1-r_2,r_2'-r_1',r_2-r_2')], \tag{A7}$$

$$G_8(r_1,r_2,r_1',r_2') = \frac{-ig^8\kappa^2}{8\pi^3}[I_8(r_1-r_2,r_1'-r_2',r_2-r_1') + I_8(r_2-r_1,r_2'-r_1',r_1-r_2')], \tag{A8}$$

$$I_4(x,y,z) = \int dkdqd\omega \frac{e^{ikx+iqy+i\omega z}J(\omega,q,k)}{(\omega-k-q+i\delta)}, \tag{A9}$$

$$I_6(x,y,z) = \int dkdqd\omega \frac{e^{ikx+iqy+i\omega z}J(\omega,q,k)(\omega-\tilde{\omega}_c-\tilde{\omega}_1)(\omega-\tilde{\omega}_c-\tilde{\omega}_2)}{(\omega-k-\tilde{\omega}_c)(\omega-q-\tilde{\omega}_c)(\omega-\tilde{\nu}_0)(\omega-\tilde{\nu}_1)(\omega-\tilde{\nu}_2)}, \tag{A10}$$

$$I_8(x,y,z) = \int dkdqd\omega \frac{e^{ikx+iqy+i\omega z}J(\omega,q,k)}{(\omega-k-\tilde{\omega}_c)(\omega-q-\tilde{\omega}_c)(\omega-\tilde{\nu}_0)(\omega-\tilde{\nu}_1)(\omega-\tilde{\nu}_2)}, \tag{A11}$$

where

$$J(\omega,q,k) = \frac{1}{(k-\tilde{\omega}_c)(\omega-k-\tilde{\omega}_1)(\omega-k-\tilde{\omega}_2)(q-\tilde{\omega}_c)(\omega-q-\tilde{\omega}_1)(\omega-q-\tilde{\omega}_2)} \tag{A12}$$

and $\tilde{\nu}_{0,1,2}$ are defined by $(\omega-\tilde{\nu}_0)(\omega-\tilde{\nu}_1)(\omega-\tilde{\nu}_2)=(\omega-\tilde{\omega}_a-\tilde{\omega}_c)[(\omega-2\tilde{\omega}_c)(\omega-\tilde{\omega}_a-\tilde{\omega}_c)-2g^2]$. The triple integrals in the definitions of $I_{4,6,8}$ are carried out analytically. It can be confirmed that the nonlinear part of the two-photon propagator, $G_{\mathrm{NL}}=-G_2(r_1-r_1')G_2(r_2-r_2')+\Sigma_{j=4,6,8}\, G_j(r_1,r_2,r_1',r_2')$, is nonzero only when the condition $\max(r_1,r_2)<\min(r_1',r_2')$ is satisfied, i.e., nonlinearity appears only after the two photons have arrived at the atom. The one- and two-photon propagators reduce to those for the one-dimensional atom in the limit of $\gamma\to 0$ and $(g,\kappa)\to\infty$, keeping $\Gamma=4g^2/\kappa$ constant [8].

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