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Nonlinear interaction of two photons with a one-dimensional atom: Spatiotemporal quantum coherence in the emitted field

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The nonlinear photon-photon interaction mediated by a single two-level atom is studied theoretically based on a one-dimensional model of the field-atom interaction. This model allows us to determine the effects of an atomic nonlinearity on the spatiotemporal coherence of a two-photon state. Specifically, the complete two-photon output wave function can be obtained for any two-photon input wave function. It is shown that the quantum interference between the components of the output state associated with different interaction processes causes bunching and antibunching in the two-photon statistics. This theory may be useful for various applications in photon manipulation, e.g., quantum information processing using photonic qubits, quantum nondemolition measurements, and the generation of entangled photons.

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I. INTRODUCTION

The nonlinearity of atomic objects, e.g., two-level atoms and quantum dots, can be sensitive to individual photons. This kind of nonlinearity may be useful for the study of photon manipulations such as quantum information processing [1–4], quantum nondemolition measurements [5], and the generation of entangled photons [6]. Realizations of single-atom nonlinearities have been studied extensively in the field of cavity quantum electrodynamics [7–9]. The sensitivity of this atom-cavity system to individual photons has been demonstrated by Turchette et al. [2]. Recently, we proposed a modification of this setup that enhances the nonlinearity by avoiding all losses in a one-sided cavity geometry [4]. Every input photon will then be found in the output. The nonlinear response of a single atom to an input of two photons, e.g., from single-photon sources [10], can then be observed in the correlations between the two output photons. In order to apply the correlations due to the nonlinear photon-photon interaction, e.g., in quantum information processing, it is important to understand the precise spatiotemporal coherence of the input and output photons. This cannot be fully achieved by theories that eliminate the quantum state of the field outside the atom-cavity system [7–9]. We therefore propose a theory that includes the propagation to and from the system in the quantum state, based on a one-dimensional model of the field-atom interaction [11]. In this paper, we apply this one-dimensional model of the field-atom interaction to the case of two-photon input wave packets.

The rest of this paper is organized as follows. In Sec. II, we give a theoretical model of the light-atom interaction in one-dimensional free space. In Sec. III, we discuss the experimental realization. In Sec. IV, we derive the general solution of the Schrödinger equation for the one-photon case. In Sec. V, we apply the result of the one-photon case to derive the general solution for two photons. In Sec. VI, the analysis of spatiotemporal coherence in the outgoing wave packet is performed as an example for the applications of our theory. In Sec. VII, the effects of the nonlinear interaction on the second-order correlations are discussed. In Sec. VIII, it is shown that the bunching and antibunching effects in the two-photon statistics can be understood as quantum interference effects of different output components. In Sec. IX, we conclude with a summary of our discussions.

II. THEORETICAL MODEL OF THE LIGHT-ATOM INTERACTION IN ONE-DIMENSIONAL FREE SPACE

To investigate the change of the spatiotemporal quantum coherence originating from the nonlinear interaction of two photons mediated by a single two-level system, we need a model for the spatiotemporal propagation to and from the atom. A possible model has been studied in the analysis of spatiotemporal quantum coherence for the case of spontaneous emission from a single excited atom [11]. This model is illustrated in Fig. 1. The $r$-axis represents the single spatial coordinate of the one-dimensional light field. A single two-level atom is coupled locally with the light field at the position $r=0$. The negative region $r<0$ and the positive region $r>0$ correspond to the incoming field and the outgoing field, respectively. This means that the light field can only propagate in the positive direction, approaching the atom at $r<0$, and moving away from it at $r>0$. The dispersion relation describing the field dynamics is then given by the wave number multiplied by the speed of light, $\omega=ck$. The Hamiltonian composed of the uninterrupted propagation and the interaction between the atom and the one-dimensional field can be written as

$$\hat{H} = \int_{-\infty}^{\infty} dk c \left[ \hat{b}^\dagger(k) \hat{b}(k) + \frac{\epsilon I}{\pi} \hat{b}^\dagger(k) \hat{a}^- - \hat{a}^- \hat{b}(k) \right], \quad (1)$$

where $\hat{b}(k)$ is the photon annihilation operator, and $\hat{a}^-$ is the annihilation operator of the atomic excitation. $\sqrt{c I/\pi}$ is the
coupling constant between the atom and the light field. $\Gamma$ is the dipole relaxation rate. As will be seen later, this rate defines the only relevant time scale of our model. Note that the Hamiltonian has been formulated in a rotating frame defined by the transition frequency $\omega_0$. Likewise, the wave vector $k$ is defined in the rotating frame, i.e., $k$ is defined relative to the resonant wave vector $\omega_0/c$.

### III. EXPERIMENTAL REALIZATION

The situation described by the theoretical model in Sec. II can be realized experimentally by using a one-sided cavity as illustrated in Fig. 2. The left mirror of the cavity has a transmittance much higher than the right mirror, which has nearly 100% reflectance. The negative region on the space axis in the model shown in Fig. 1 corresponds to the input in Fig. 2 and the positive region corresponds to the output in Fig. 2.

In terms of the conventional cavity quantum electrodynamics parameters, this regime is characterized by $\kappa \gg g$, where $\kappa$ is the cavity damping rate through the left mirror and $g$ is the dipole coupling between the atom and the cavity mode. Therefore the method of adiabatic elimination can be applied to the time evolution of the cavity field [7]. This means that since the cavity damping rate $\kappa$ is much faster than the dipole coupling $g$, the interaction between the atom and the outside field mediated by the cavity field can be expressed by an effective dipole relaxation rate $\Gamma = g^2/\kappa$. It should be noted that, in this case, Hamiltonian (1) represents an approximation valid only within the finite cavity bandwidth of $2\kappa$. Effectively, the theoretical model can then be used to correctly describe the atom-cavity dynamics at time scales larger than $1/\kappa$. This approximation is sufficient for the description of atomic absorption and emission processes as long as $1/\Gamma \gg 1/\kappa$. Features of the cavity response which become obvious only at a time scale of about $1/\kappa$ are neglected in this paper.

The dipole relaxation rate $\Gamma$ describes the dipole damping caused by emissions through the left mirror of the cavity, and the corresponding rate of spontaneous emission through the cavity is equal to $2\Gamma$ [4,7,9]. In our case, we assume that the rate of spontaneous emission into the noncavity modes $\gamma_i$ is negligible ($\gamma_i \ll 2\Gamma$). Nearly all emissions from the atom can then be confined to the cavity and $2\Gamma$ is the total spontaneous emission rate of the excited atom in the cavity. In present cavity designs, this can be realized by covering a large solid angle of the atomic emission with the cavity mirrors and exploiting the enhancement of spontaneous emission by the cavity. For example, in the experiment of Turchette et al. [2], the cavity parameters indicate that about 70% of the spontaneous emission from the atom is emitted through the single cavity mode. Another promising method of achieving a one-dimensional emission and absorption of the atom is the use of semiconductor microstructures, as reported, e.g., in Ref. [12]. In any case, our model should apply to any cavity design with $\kappa \gg g \gg \gamma_i$.

### IV. ONE-PHOTON PROCESSES

In this section, we treat the interaction of one photon with the atomic system as a preparation for the analysis of two-photon processes. We can expand the quantum state of the single photon in the basis of the wave-number eigenstates $|k\rangle$ and the excited state $|E\rangle$ of the two-level atom. The quantum state for the one-photon process can then be written as

$$|\Psi(t)\rangle = |E; t\rangle|E\rangle + \int_{-\infty}^{\infty} dk \Psi(k; t)|k\rangle. \quad (2)$$

In this basis, The Hamiltonian given by Eq. (1) can be expressed as

$$\hat{H}_{\text{1photon}} = \hbar c \hat{k} + \hat{H}_{\text{int}}, \quad (3)$$

with $\hat{k} = \int_{-\infty}^{\infty} dkk|k\rangle\langle k|$ and $\hat{H}_{\text{int}}$

$$= i\hbar \sqrt{\frac{\Gamma}{\pi}} \int_{-\infty}^{\infty} dk (|k\rangle\langle E| - |E\rangle\langle k|).$$
The equations for the time evolution of the probability amplitudes \( \Psi(E;t) \) and \( \Psi(k;t) \) can then be obtained from the Schrödinger equation \( i\hbar \frac{d}{dt} |\Psi(t)\rangle = \hat{H} |\Psi(t)\rangle \) using Eqs. (2) and (3),

\[
\frac{d}{dt} \Psi(E;t) = -\sqrt{\frac{\Gamma}{\pi}} \int_{-\infty}^{\infty} dk \Psi(k;t),
\]

\[
\frac{d}{dt} \Psi(k;t) = -i c \Psi(k;t) + \sqrt{\frac{\Gamma}{\pi}} \Psi(E;t).
\]

The time evolution \( \Psi(k;t) \) can be obtained by integrating Eq. (5).

\[
\Psi(r;t) = \begin{cases} 
\Psi(r-c(t-t_i);t_i) & \text{for } r<0 \text{ or } c(t-t_i)<r, \\
\Psi(r-c(t-t_i);t_i) + \sqrt{\frac{2\Gamma}{c}} \Psi(E;t - \frac{r}{c}) & \text{for } 0<r<c(t-t_i).
\end{cases}
\]

The top term corresponds to the single-photon amplitude propagating without being absorbed by the atom. On the other hand, the bottom term consists of two processes. The first term also corresponds to propagation without absorption, while the second term corresponds to the amplitude of a single photon reemitted into the outgoing field after absorption by the atom [11]. The time evolution \( \Psi(E;t) \) of the excited-state amplitude can be obtained by integrating Eq. (4) using the result for \( \Psi(k;t) \) given in Eq. (6),

\[
\Psi(E;t) = -\sqrt{\frac{\Gamma}{\pi}} \int_{t_i}^{t} dt' \int_{-\infty}^{\infty} dk \Psi(k;t)
\]

\[
= -\sqrt{\frac{\Gamma}{\pi}} \int_{t_i}^{t} dt' \int_{-\infty}^{\infty} dk \left( e^{-i k c(t-t_i)} \Psi(k;t_i) + \sqrt{\frac{c}{2\Gamma}} \int_{t_i}^{t} dt' e^{-i k c(t-t')} \Psi(E;t') \right).
\]

Using the Fourier transform to obtain the real-space representation of \( \Psi(k;t) \), the result reads

\[
\Psi(r;t) = \begin{cases} 
\Psi(r-c(t-t_i);t_i) & \text{for } r<0, \\
\Psi(r-c(t-t_i);t_i) - 2\Gamma \int_{t_i}^{t} dt' e^{-\Gamma(t-t_i-t')} \Psi(-c(t'-t_i);t_i) & \text{for } 0<r<c(t-t_i), \\
0 & \text{for } r>c(t-t_i).
\end{cases}
\]
To investigate the outgoing amplitude $\Psi(r>0; t)$ for an arbitrary incoming amplitude under conditions (11) and (12), it is convenient to represent $\Psi(r>0; t)$ by using the matrix element of the time evolution operator. In order to derive this matrix element, we expand the time evolution $|\Psi(t)\rangle$ of the quantum state given by Eqs. (11) and (12) as follows:

$$|\Psi(t)\rangle = \hat{U}(t-t_i)|\Psi(t_i)\rangle = \int_{-\infty}^{\infty} dr dr' |r\rangle \langle r| \hat{U}(t-t_i)|r'\rangle \langle r'| |\Psi(t_i)\rangle,$$

$$+ |E\rangle \int_{-\infty}^{\infty} dr' \langle E| \hat{U}(t-t_i)|r'\rangle \langle r'| |\Psi(t_i)\rangle,$$

$$|\Psi(t)\rangle = \int_{-\infty}^{\infty} dr dr' |r\rangle \langle r| \hat{U}(t-t_i)|r'\rangle \langle r'| |\Psi(t_i)\rangle$$

where $|r\rangle = (1/\sqrt{2\pi}) \int_{-\infty}^{\infty} dk e^{-ikr}|k\rangle$ is the eigenstate of a photon at the position $r$. $\hat{U}(t-t_i) = e^{-(i\hbar)(t-t_i)}$ is the time evolution operator. $\Psi(r,t)$ can then be expressed as follows:

$$\Psi(r,t) = \langle r| \Psi(t)\rangle = \int_{-\infty}^{\infty} dr' u_{1\text{photon}}(r,r'; t-t_i) \Psi(r'; t_i),$$

where

$$u_{1\text{photon}}(r,r'; t-t_i) = \langle r| \hat{U}(t-t_i)|r'\rangle.$$
be expressed using the single-photon Hamiltonian
\[ \hat{H}^{(i)}_{\text{1photon}} \] and the matrix element of the
interaction free dynamics is then given by the
Hamiltonian including transitions to the state \( \langle E_i | \rangle \).

This Hamiltonian describes the interaction free evolution of the
two-photon state. Therefore the time evolution of the
two-photon probability amplitudes \( \Psi^{\text{lin}}(k_1,k_2;t) \), \( \Psi^{\text{lin}}(E_1,k_2;t) \), and \( \Psi^{\text{lin}}(k_1,E_2;t) \) given by this Hamiltonian
corresponds to the results of the one-photon case given in
Eqs. (4)–(5). It reads
\[
\frac{d}{dt} \Psi^{\text{lin}}(k_1,k_2;t) = -i c (k_1 + k_2) \Psi^{\text{lin}}(k_1,k_2;t) \\
+ \sqrt{\frac{c \Gamma}{\pi}} \left[ \Psi^{\text{lin}}(k_1,E_2;t) \\
+ \Psi^{\text{lin}}(E_1,k_2;t) \right],
\]
(20)
\[
\frac{d}{dt} \Psi^{\text{lin}}(E_1,k_2;t) = -i c k_2 \Psi^{\text{lin}}(E_1,k_2;t) \\
- \sqrt{\frac{c \Gamma}{\pi}} \int_{-\infty}^{\infty} dk_1 \Psi^{\text{lin}}(k_1,k_2;t) \\
+ \sqrt{\frac{c \Gamma}{\pi}} \Psi^{\text{lin}}(E_1,E_2;t),
\]
(21)
\[
\frac{d}{dt} \Psi^{\text{lin}}(k_1,E_2;t) = -i c k_1 \Psi^{\text{lin}}(k_1,E_2;t) \\
- \sqrt{\frac{c \Gamma}{\pi}} \int_{-\infty}^{\infty} dk_2 \Psi^{\text{lin}}(k_1,k_2;t) \\
+ \sqrt{\frac{c \Gamma}{\pi}} \Psi^{\text{lin}}(E_1,E_2;t).
\]
(22)

Since the time evolution described by \( \hat{H}^{\text{lin}} \) corresponds to the
time evolution of the single photon case, the integration can be
performed according to the procedure in Eqs. (6)–(10), and the matrix element \( \mathbf{u}^{\text{lin}}_{\text{2photon}}(r_1,r_2;r'_1,r'_2;t-t_i) \) of the
time evolution operator can be expressed as a product of the
individual single-photon matrix elements given by Eq. (16),
\[
\mathbf{u}^{\text{lin}}_{\text{2photon}}(r_1,r_2;r'_1,r'_2;t-t_i)
= \mathbf{u}_{\text{1photon}}(r_1;r'_1;t-t_i) \cdot \mathbf{u}_{\text{1photon}}(r_2;r'_2;t-t_i).
\]
(23)
The photon-photon interaction can now be included by adding the nonlinear term that suppresses transitions to the two-photon absorption state $|E_1, E_2\rangle$, 
\[\hat{H}_{\text{2phot}} = \hat{H}_{\text{lin}} + \Delta \hat{H}_{\text{Nonlin}},\]
where \[\Delta \hat{H}_{\text{Nonlin}} = -(\hat{F}_{\text{int}}^{(1)} \otimes |E_2\rangle \langle E_2|) + |E_1\rangle \langle E_1| \otimes \hat{H}_{\text{int}}^{(2)}).\] (24)

With this addition, the matrix elements of the two-photon Hamiltonian (24) are identical with the corresponding two-photon matrix elements of the original Hamiltonian (1). The time evolution of the wave function is modified only slightly by the addition of the nonlinear term. It now reads
\[
\frac{d}{dt} \Psi(k_1,k_2;t) = -ic(k_1+k_2) \Psi(k_1,k_2;t) + \sqrt{\frac{c^2 \Gamma}{\pi}} \left[ \Psi(k_1, E_2; t) + \Psi(E_1, k_2; t) \right],
\]
(25)
\[
\frac{d}{dt} \Psi(E_1,k_2;t) = -ic k_2 \Psi(E_1,k_2;t) + \sqrt{\frac{c^2 \Gamma}{\pi}} \int_{-\infty}^{\infty} dk_1 \Psi(k_1,k_2;t),
\]
(26)
\[
\frac{d}{dt} \Psi(k_1,E_2;t) = -ic k_1 \Psi(k_1,E_2;t) + \sqrt{\frac{c^2 \Gamma}{\pi}} \int_{-\infty}^{\infty} dk_2 \Psi(k_1,k_2;t).
\]
(27)

The comparison between Eqs. (21)–(22) and Eqs. (26)–(27) shows that the integration of Eqs. (26) and (27) is the same as the integration of Eqs. (21) and (22), except that $\Psi(E_1, E_2; t)$ is zero in the integration of $\Psi(E_1, k_2; t)$ and $\Psi(k_1, E_2; t)$. This means that the matrix element $u_{\text{2phot}}^{(t_1, t_2; r_1', r_2'; t-t_i)}$ of the time evolution described by $\hat{H}_{\text{2phot}}$ can be obtained by identifying the components in the interaction free propagation of the two photons from $r_1', r_2'$ to $r_1, r_2$ described by the matrix element $u_{\text{lin}}^{(t_1, t_2; r_1', r_2'; t-t_i)}$ and removing the ones due to two-photon absorption.

For the case that photon 1 and photon 2 start at $r_2' < r_1' < 0$, the time evolution of this probability amplitude can be obtained by distinguishing the following three time regions.

*Time region I, $t < t_i - r_1'/c$: the two photons are independently propagating in the incoming field [see Fig. 4(a)]. The matrix element associated with the uninterrupted propagation $u_{\text{2phot}}^{(t_1, t_2; r_1', r_2'; t-t_i)}$ can be expressed as
\[
 u_{\text{2phot}}^{(t_1, t_2; r_1', r_2'; t-t_i)} = u_{\text{prop}}^{(r_1'; t-t_i)} \cdot u_{\text{prop}}^{(r_2'; t-t_i)}. \]
(28)

*Time region II, $t_i - r_1'/c < t < t_i - r_2'/c$: photon 1 has already arrived at the atom and photon 2 has not arrived yet. In this situation, photon 1 is interacting with the atom and photon 2 is propagating in the incoming field [see Fig. 4(b)]. The matrix element associated with one photon absorption $u_{\text{phot}}^{(t_1, t_2; r_1', r_2'; t-t_i)}$ can be expressed as
\[
 u_{\text{phot}}^{(t_1, t_2; r_1', r_2'; t-t_i)} = u_{\text{prop}}^{(r_1'; t-t_i)} \cdot u_{\text{prop}}^{(r_2'; t-t_i)} \cdot u_{\text{abs}}^{(c(t-t_i) + r_1', r_2')}.
\]
(29)

*Time region III, $t_i - r_2'/c < t$: includes a new situation not treated in the one-photon processes, because both photons have now reached the atom. In this time region, the time evolution given by $\hat{H}_{\text{2phot}}$ is different from the one given by $\hat{H}_{\text{int}}$ because there cannot be any contributions of the two-photon absorption amplitude $\Psi(E_1, E_2; t-t_i)$ in the time...
evolution of the outgoing amplitude. This means that the matrix element corresponding to two-photon absorption \( \mathbf{u}_{\text{abs}}(r_1; r'_1; t - t_i) \cdot \mathbf{u}_{\text{abs}}(r_2; r'_2; t - t_i) \) will not be included in the time evolution of \( \mathbf{u}_{\text{2phot}}(r_1; r_2; r'_1; r'_2; t - t_i) \) if the position \( r_1 \) of photon 1 is less than \( c(t - t_i) + r'_2 \), hence, in this case, photon 2 arrives at the atom before photon 1 is reemitted. The matrix element \( \mathbf{u}_{\text{2phot}}^{\text{III}}(r_1; r_2; r'_1; r'_2; t - t_i) \) can then be expressed as

\[
\mathbf{u}_{\text{2phot}}^{\text{III}}(r_1; r_2; r'_1; r'_2; t - t_i) = \begin{cases} 
\mathbf{u}_{\text{1photon}}(r_1; r'_1; t - t_i) \cdot \mathbf{u}_{\text{1photon}}(r_2; r'_2; t - t_i) & \text{for } r_1 > c(t - t_i) + r'_2, \\
\mathbf{u}_{\text{1photon}}(r_1; r'_1; t - t_i) \cdot \mathbf{u}_{\text{prop}}(r_2; r'_2; t - t_i) & \text{for } r_1 < c(t - t_i) + r'_2.
\end{cases}
\]

Note that the dependence of the transition amplitudes for photon 2 on the output coordinate of photon 1 makes it impossible to separate the dynamics of photon 1 and photon 2. Therefore, it is not possible to illustrate this time region using single-photon amplitudes.

The matrix element with \( r'_2 < r'_1 \) can be obtained from the results for \( r'_2 < r'_1 \) by using the positive symmetry between photons propagating in free space. The results of these time regions can be summarized as follows:

\[
\mathbf{u}_{\text{2phot}}(r_1; r_2; r'_1; r'_2; t - t_i) = \mathbf{u}_{\text{2phot}}^{\text{lin}}(r_1; r_2; r'_1; r'_2; t - t_i) + \Delta \mathbf{u}_{\text{2phot}}^{\text{Nonlin}}(r_1; r_2; r'_1; r'_2; t - t_i),
\]

\[
\mathbf{u}_{\text{2phot}}^{\text{lin}}(r_1; r_2; r'_1; r'_2; t - t_i) = \mathbf{u}_{\text{1photon}}(r_1; r'_1; t - t_i) \cdot \mathbf{u}_{\text{1photon}}(r_2; r'_2; t - t_i),
\]

\[
\Delta \mathbf{u}_{\text{2phot}}^{\text{Nonlin}}(r_1; r_2; r'_1; r'_2; t - t_i) = - \frac{4 \Gamma^2}{c^2} e^{-(\sqrt{\Gamma/c})r'_1 + 2r(t - t_i) - r'_1 - r_2}
\]

for \( 0 < r_1, r_2 < c(t - t_i) + \text{min}(r'_1, r'_2) \),

where \( \text{min}(r'_1, r'_2) \) is the minimum of \( r'_1 \) and \( r'_2 \). The matrix element \( \Delta \mathbf{u}_{\text{2phot}}^{\text{Nonlin}} \) of the nonlinear interaction between the two photons originates from the impossibility of two-photon absorption at the single two-level atom. The remainder of the dynamics is identical to the single-photon processes. The total output wave function of two photons propagating in the one-dimensional field can then be expressed as

\[
\Psi(r_1, r_2; t) = \int_{-\infty}^{\infty} dr'_1 dr'_2 \mathbf{u}_{\text{2phot}}(r_1; r_2; r'_1; r'_2; t - t_i) \cdot \Psi(r'_1, r'_2; t). 
\]

(32)

The output wave function describes the state of the photons propagating in the far field after the interaction with the atom. In general, a two-photon wave function propagating in one-dimensional free space obeys the relation \( \Psi(r_1, r_2; t) = \Psi(r_1 - ct, r_2 - ct; 0) \). Therefore, the function of three parameters \( \Psi(r_1, r_2; t_i) \) can be expressed by the function of two parameters \( \Psi_{\text{in}}(r_1 - ct, r_2 - ct; t_f) \). In the same way, the two-photon wave function \( \Psi(r_1, r_2; t_f) \) in the outgoing far field can be expressed as \( \Psi_{\text{out}}(r_1 - ct_f, r_2 - ct_f) \). We can then simplify our result (32) by the transformation to a moving coordinate system,

\[
r_1 - ct = x_1, \\
r_2 - ct = x_2.
\]

(33)

In this coordinate system, the output state at an arbitrary time \( t_f \) can be expressed as

\[
\Psi_{\text{out}}(x_1, x_2) = \Psi(r_1, r_2; t_f) = \int_{-\infty}^{\infty} dx'_1 dx'_2 \mathbf{u}(x_1, x_2; x'_1, x'_2) \cdot \Psi_{\text{in}}(x'_1, x'_2),
\]

with \( x_1 = r_1 - ct_f, \quad x_2 = r_2 - ct_f, \quad x'_1 = r'_1 - ct_f, \quad \text{and} \quad x'_2 = r'_2 - ct_f \),

(34)

where \( \mathbf{u}(x_1, x_2; x'_1, x'_2) \) is given by

\[
\mathbf{u}(x_1, x_2; x'_1, x'_2) = \mathbf{u}_{\text{lin}}(x_1, x_2; x'_1, x'_2) + \Delta \mathbf{u}_{\text{Nonlin}}(x_1, x_2; x'_1, x'_2),
\]

(35)

where \( \mathbf{u}_{\text{lin}}(x_1, x_2; x'_1, x'_2) = \mathbf{u}_{\text{1photon}}(x_1; x'_1) \cdot \mathbf{u}_{\text{1photon}}(x_2; x'_2), \)

(36)

with \( \mathbf{u}_{\text{1photon}}(x; x') = \delta(x - x') - \frac{2 \Gamma}{c} e^{-(\sqrt{\Gamma/c})(x' - x)} \)

for \( x \approx x' \)

(37)

and \( \Delta \mathbf{u}_{\text{Nonlin}}(x_1, x_2; x'_1, x'_2) = - \frac{4 \Gamma^2}{c^2} e^{-(\sqrt{\Gamma/c})(x'_1 + x'_2 - x_1 - x_2)} \)

for \( x_1, x_2 < \text{min}(x'_1, x'_2) \).

(38)

Note that the linear component \( \mathbf{u}_{\text{lin}}(x_1, x_2; x'_1, x'_2) \) of the matrix element \( \mathbf{u}(x_1, x_2; x'_1, x'_2) \) represents the time evolution of the two photons without interaction. Therefore it can be ex-
pressed by the product of two one-photon matrix elements. On the other hand, the nonlinear component \( \Delta u^{\text{Nonlin}}(x_1, x_2) \) represents the difference from the time evolution of the linear component. The output wave function \( \Psi_{\text{out}}(x_1, x_2) \) can then be expressed as the superposition of the linear term and the nonlinear term by substituting the right hand side of Eq. (35) into Eq. (34),

\[
\Psi_{\text{out}}(x_1, x_2) = \Psi_{\text{lin}}(x_1, x_2) + \Delta \Psi_{\text{out}}^{\text{Nonlin}}(x_1, x_2),
\]

(39)

\[
\Psi_{\text{lin}}(x_1, x_2) = \int_{-\infty}^{\infty} dx_1' dx_2' \Psi_{\text{in}}(x_1', x_2') \Psi_{\text{lin}}(x_1', x_2),
\]

(40)

\[
\Delta \Psi_{\text{out}}^{\text{Nonlin}}(x_1, x_2) = \int_{-\infty}^{\infty} dx_1' dx_2' \Delta u^{\text{Nonlin}}(x_1, x_2; x_1', x_2') \Psi_{\text{lin}}(x_1', x_2).
\]

(41)

The formulation above can be used to analyze the outgoing amplitude for any arbitrary incoming two-photon state. In the following section, these results are applied to the case of rectangular two-photon input wave packets.

**VI. GENERAL SOLUTION FOR RECTANGULAR INPUT WAVE PACKETS**

In order to investigate the typical properties of the nonlinear photon-photon interaction, a rectangular two-photon input wave packet is convenient because the homogeneous probability distribution of the two input photons makes it easier to understand the change of the correlations between the two photons due to the interaction. We assume that the shape of a single two-photon pulse prepared in the incoming far field is a rectangle of length \( L \). Such a rectangular two-photon wave packet can be written as follows:

\[
\Psi_{\text{in}}(x_1, x_2) = \Psi_{\text{in}}(x_1) \cdot \Psi_{\text{in}}(x_2),
\]

(42)

\[
\Psi_{\text{in}}(x) = \begin{cases} \sqrt{\frac{2}{\pi L}} e^{-x^2/(2L^2)} & \text{for } 0 \leq x \leq L, \\ 0 & \text{otherwise.} \end{cases}
\]

(43)

Note that, in any practical situation, the flanks of a rectangular pulse will not rise and fall infinitely fast. The discontinuities of the wave function should therefore be interpreted as a continuous change of amplitude that is extremely fast on a time scale of \( 1/\Gamma \). In particular, the flanks of the pulse should be smooth at a timescale of \( 1/\kappa \) due to the limitations of the model regarding the description of the cavity dynamics (see Sec. III). However, we assume this timescale to be so much shorter than \( 1/\Gamma \) that its effects can be neglected in the following.

As shown in Eq. (39), the output wave packet can be separated into a linear term and a nonlinear term. The linear term in the output of the rectangular input wave packet described by Eq. (42) can be obtained according to Eq. (40). It reads

\[
\Psi_{\text{out}}(x_1, x_2) = \int_{-\infty}^{\infty} dx_1' dx_2' \Psi_{\text{in}}(x_1', x_2') \Psi_{\text{lin}}(x_1', x_2) = \Psi_{\text{lin}}(x_1) \cdot \Psi_{\text{lin}}(x_2).
\]

(44)

The nonlinear term in the output of the rectangular input wave packet described by Eq. (42) can be obtained according to Eq. (41). It reads

\[
\Delta \Psi_{\text{out}}^{\text{Nonlin}}(x_1, x_2) = \int_{-\infty}^{\infty} dx_1' dx_2' \Delta u^{\text{Nonlin}}(x_1, x_2; x_1', x_2') \Psi_{\text{lin}}(x_1', x_2).
\]

(45)

\[
\Psi_{\text{out}}(x_1, x_2) = \Psi_{\text{lin}}(x_1, x_2) + \Delta \Psi_{\text{out}}^{\text{Nonlin}}(x_1, x_2).
\]

(46)

It should be noted that the shape of the output wave packet is defined by the ratio of the dipole relaxation length \( c/\Gamma \) and the length \( L \) of the input wave packet. In frequency representation, this means that the shape of the output wave packet is sensitive to whether the frequency spectrum of the input wave packet is narrower than the atomic line width \( 2\Gamma \) or not. A particularly simple case for showing the contribution of the nonlinear term to the output wave packet can be obtained in the long pulse limit \( c/\Gamma \ll L \) because, in this limit, the linear term given by Eq. (43) of the output wave packet is almost constant for the region \( 0 < x_i < L - 2c/\Gamma (i = 1, 2) \), that is,

\[
\Psi_{\text{lin}}^{\text{out}}(x_1, x_2) = \frac{1}{L} \Psi_{\text{lin}}(x_1, x_2) \quad \text{for } 0 < x_i < L - 2c/\Gamma \quad (i = 1, 2).
\]

(47)
which only depends on the relative distance \( |x_1 - x_2| \) between the two photons. The exponential decay indicates that the nonlinear deviation from the linear term only becomes significant in the vicinity of \( x_1 = x_2 \). The output wave function can then be written as

\[
\Psi_{\text{out}}(x_1, x_2) = \frac{1}{L} \frac{4}{L} e^{-(G/\Gamma)} |x_1 - x_2|
\]

for \( 0 < x_i < L - 2c/\Gamma \) \( (i = 1, 2) \). (48)

The contour plot in Fig. 5(a) shows an example of the output wave function \( \Psi_{\text{out}}(x_1, x_2) \) in the long pulse limit. The probability amplitude increases from black to white shading. In this example, we have chosen an input wave packet length \( L = 20c/\Gamma \), which is 20 times greater than dipole relaxation length \( c/\Gamma \). The plateau region away from \( x_1 = x_2 \) in Fig. 5(a) has a positive amplitude. On the other hand, in the vicinity of \( x_1 = x_2 \), a valley of negative amplitude cuts across this plateau. The shape of this valley can be seen in Fig. 5(b). Figure 5(b) is the cross section of the contour plot at \( x_2 = 10c/\Gamma \). It should be noted that the shape of the valley is the same for any cross section \( x_2 \) within the plateau region. The valley is due to the contribution of the nonlinear term in Eq. (44) which decreases with the distance between \( x_1 \) and \( x_2 \). The plateau is the unchanged characteristic feature of the long rectangular input wave packet. Figure 5 shows the typical characteristics in the output for a long two-photon input pulse. In the following section, these typical characteristics are analyzed in terms of two-photon statistics.

VII. ANALYSIS OF TWO-PHOTON STATISTICS

Two-photon statistics are often used to characterize the properties of nonclassical light from the viewpoint of photon counting. For this purpose, the second-order correlation

\[
G_{\text{out}}^{(2)}(t, t + \tau)
\]

is defined by the joint probability density of detecting one photon at time \( t + \tau \) when the other photon is detected at time \( t \). When the second-order correlation \( G_{\text{out}}^{(2)}(t, t + \tau) \) is expressed by using the probability density \( |\Psi_{\text{out}}(x + c\tau, t)|^2 \), the time \( t \) is given by the space coordinate \( x \) divided by the speed of light, \( t = x/c \). Also, the probability-density \( |\Psi_{\text{out}}(x + c\tau, t)|^2 \) must be rescaled by a factor of \( c^2 \), since the probability density per unit time is expressed by \( c^2 \) times the probability density per unit length and, for second-order correlations, this factor is squared. Moreover, \( G_{\text{out}}^{(2)} \) does not distinguish between the two photons. Therefore, both \( |\Psi_{\text{out}}(x, x + c\tau)|^2 \) and \( |\Psi_{\text{out}}(x + c\tau, t)|^2 \) contribute to \( G_{\text{out}}^{(2)}(t, t + \tau) \). Taking all these factors into account, the second-order correlation function can be written as

\[
G_{\text{out}}^{(2)}(-x/c, -x/c + \tau) = c^2 \left[ |\Psi_{\text{out}}(x + c\tau, t)|^2 + |\Psi_{\text{out}}(x, x + c\tau)|^2 \right]
\]

\[
= 2c^2 |\Psi_{\text{out}}(x + c\tau, t)|^2. \tag{49}
\]

Note that \( |\Psi_{\text{out}}(x + c\tau, t)|^2 \) and \( |\Psi_{\text{out}}(x, x + c\tau)|^2 \) always have to be equal to each other because of the bosonic nature of the two photons. The contour plot of the second-order correlations is shown in Figs. 6(a-1) and 6(a-2) for an input pulse length of \( L = 20c/\Gamma \). The correlation increases from black to white shading. To indicate the contribution of the nonlinear term described by Eq. (44) in the second-order correlations, it is convenient to compare this result with the second-order correlations obtained from the linear component of the output only. Figures 6(b-1) and 6(b-2) shows this second-order correlation \( G_{\text{out}}^{(2) \text{lin}}(-x/c, -x/c + \tau) = 2c^2 |\Psi_{\text{out}}^{\text{lin}}(x + c\tau, t)|^2 \) of the linear component. The comparison of these figures shows that both cases are identical except for their distributions around \( \tau = 0 \). As can be seen in Fig. 6(a-2), the nonlinear interaction causes photon bunching.
around $\tau=0$ and photon antibunching around $\tau = \pm 2 \ln 2/1$. In order to compare the second-order correlations with other systems in quantum optics, it is useful to normalize the correlation function $G_{\text{out}}^{(2)}$ by the product of the probability densities of single-photon detection at times $-x/c$ and $-x/c + \tau$ in the output field. In the long pulse limit, the total probability of finding both photons within the output region of $0 < x < L (i=1,2)$ is nearly equal to 1. Therefore the single-photon detection probability density per unit time within this region is $2c/L$. By using this average photon density, we obtain the normalized second-order correlation function,

$$g_{\text{out}}^{(2)}(-x/c, -x/c + \tau) = G_{\text{out}}^{(2)}(-x/c, -x/c + \tau)/(2c/L)^2$$

$$= \frac{L^2}{2} |\Psi_{\text{out}}(x+c, x, x)|^2. \quad (50)$$

With the approximations for the long pulse limit given by Eq. (48), this correlation reads

$$g_{\text{out}}^{(2)}(-x/c, -x/c + \tau) = \frac{1}{2} (1 - 4e^{-\Gamma|\tau|})^2$$

for $0 \leq \{x+c, x\} \leq L-2c/\Gamma$. \quad (51)
For $x_1 < x_1'$ and $x_2 < x_2'$, the components of $u(x_1, x_2; x_1', x_2')$ read

$$u^{(i)}(x_1, x_2; x_1', x_2') = u_{\text{prop}}(x_1; x_1') \cdot u_{\text{prop}}(x_2; x_2'),$$

$$u^{(ii)}(x_1, x_2; x_1', x_2') = u_{\text{prop}}(x_1; x_1') \cdot u_{\text{abs}}(x_2; x_2') + u_{\text{abs}}(x_1; x_1') \cdot u_{\text{prop}}(x_2; x_2'),$$

$$u^{(iii)}(x_1, x_2; x_1', x_2') = u_{\text{abs}}(x_1; x_1') \cdot u_{\text{abs}}(x_2; x_2') + \Delta \Psi^{(ii)}(x_1, x_2; x_1', x_2').$$

Therefore, the output wave packet $\Psi^{(ii)}(x_1, x_2)$ can be expanded as

$$\Psi^{(ii)}(x_1, x_2) = \Psi^{(i)}(x_1, x_2) + \Psi^{(ii)}(x_1, x_2) + \Psi^{(iii)}(x_1, x_2),$$

where $\Psi^{(i)-(iii)}$ is given by

$$\Psi^{(i)-(iii)}(x_1, x_2) = \int_{-\infty}^{\infty} dx_1' dx_2' \Psi^{(i)-(iii)}(x_1, x_2; x_1', x_2') \cdot \Psi_m(x_1', x_2').$$

For the rectangular input wave packet, the output wave function in the interval $0 < x_i < L$ ($i = 1, 2$) is given by

$$\Psi^{(i)}(x_1, x_2) = \frac{1}{L},$$

$$\Psi^{(ii)}(x_1, x_2) = \frac{1}{L} (2e^{-(\Gamma/c)(L-x_1)} - 2)$$

$$+ \frac{1}{L} (2e^{-(\Gamma/c)(L-x_2)} - 2),$$

$$\Psi^{(iii)}(x_1, x_2) = \frac{4}{L} (e^{-(\Gamma/c)(L-x_1)} - 1)(e^{-(\Gamma/c)(L-x_2)} - 1)$$

$$+ \Delta \Psi^{(ii)}(x_1, x_2).$$

Figure 8(a) shows the correlations of $\Psi^{(i)-(iii)}(x_1, x_2)$ at $x_2 = 10c/\Gamma$ for an input pulse length of $L = 20c/\Gamma$. The dotted line shows the amplitude corresponding to process (i), where both of two photons are transmitted without absorption by the atom. Likewise, the short dashed line shows the amplitude corresponding to process (ii), where one photon is absorbed and then reemitted and another photon is transmitted without absorption. The thin line shows the amplitude corresponding to process (iii), where both photons are absorbed and then reemitted. Figure 8(b) shows the superposition of all the amplitudes. This superposition is equal to the output amplitude $\Psi^{(ii)}(x_1, x_2)$ for $x_2 = 10c/\Gamma$. Therefore, Fig. 8(b) is identical with Fig. 5(b). Note that the discontinuities of the output wave function $\Psi^{(ii)}(x_1, x_2)$ originate from the direct transmission of the rectangular input wave packet in $\Psi^{(ii)}$. As mentioned in Sec. VI, these discontinuities represent changes of the amplitude that are extremely fast on a time scale of $1/\Gamma$, but would be continu-
corresponds to the difference between the detection times of the photons at the hand, when the relative distance is much smaller than the dipole relaxation length. The distribution only depends on the relative distance. The thin line shows the amplitude corresponding to process (iii), where both photons are absorbed and then reemitted. The superposition of all the amplitudes is shown in (b). Note that (b) is identical to Fig. 5(b).

Fig. 8. Analysis of the interaction processes of two photons and the atom for an input pulse length of \( L = 20c/\Gamma \). In (a), the dotted line shows the amplitude at \( x_2 = 10c/\Gamma \) corresponding to process (i), where both photons are transmitted without absorption. The short dashed line shows the amplitude corresponding to process (ii), where one photon is absorbed and then reemitted while the other photon is transmitted without absorption. The thin line shows the amplitude corresponding to process (iii), where both photons are absorbed and then reemitted. The superposition of all the amplitudes is shown in (b). With that (b) is identical to Fig. 5(b).

To understand the details of the two time correlations, we now examine the effect of the nonlinear contribution \( \Delta \Psi_{\text{out}}^{\text{Nonlin}}(x_1,x_2) \) in the superposition. The nonlinear contribution only depends on the relative distance \( |x_2-x_1| \) which corresponds to the difference between the detection times of the two photons. When the relative distance \( |x_2-x_1| \) is much larger than the dipole relaxation length \( c/\Gamma \), the nonlinear contribution \( \Delta \Psi_{\text{out}}^{\text{Nonlin}} \) is nearly equal to zero and the double absorption amplitude \( \Psi_{\text{out}}^{(ii)} \) is close to \( 4/L \). On the other hand, when the relative distance \( |x_2-x_1| \) is much smaller than the dipole relaxation length \( c/\Gamma \), the nonlinear contribution \( \Delta \Psi_{\text{out}}^{\text{Nonlin}} \) is nearly equal to \( -4/L \) and the double absorption amplitude \( \Psi_{\text{out}}^{(ii)} \) is close to zero. As noted previously, \( \Delta \Psi_{\text{out}}^{\text{Nonlin}} \) represents the absence of simultaneous double absorption. The time dependence of \( \Delta \Psi_{\text{out}}^{\text{Nonlin}} \) therefore describes the saturation dynamics of the two-level atom. Since the no-absorption amplitude \( \Psi_{\text{out}}^{(i)} \) and the single-absorption amplitude \( \Psi_{\text{out}}^{(ii)} \) are independent of the saturation and their amplitudes are \( 1/L \) and \(-4/L\), respectively, the total amplitude \( \Psi_{\text{out}} \) is obtained by adding a constant value of \(-3/L\) to \( \Psi_{\text{out}}^{(ii)} \). The resulting total amplitude \( \Psi_{\text{out}} \) is then close to \( 1/L \) for relative distances \( |x_2-x_1| \gg c/\Gamma \) and drops to \(-3/L\) for relative distances \( |x_2-x_1| \ll c/\Gamma \). The total amplitude \(-3/L\) for relative distances \( |x_2-x_1| \ll c/\Gamma \) is associated with the bunching effect in the second-order correlations. The total amplitude then changes from positive to negative values continuously, depending on the relative distance between the two photons \( |x_2-x_1| \). Precisely speaking, the total amplitude \( \Psi_{\text{out}} \) is positive if the relative distance is \( |x_2-x_1| > 2 \ln(2)c/\Gamma \) and negative if the relative distance is \( |x_2-x_1| < 2 \ln(2)c/\Gamma \) [see Fig. 8(b)]. That is, in the region with \( |x_2-x_1| < 2 \ln(2)c/\Gamma \), the interaction of the two photons causes a phase flip of \( \pi \). This phase flip can be understood as evidence on the quantum level for the resonant nonlinearity discussed in Ref. [4]. The antibunching at \( |x_2-x_1| = 2 \ln(2)c/\Gamma \) originates from \( \Psi_{\text{out}} \) passing through zero as the sign of the total amplitude changes from negative to positive. In this way, both the bunching and the antibunching effects in the two-photon correlation can be explained in terms of interference effects between the quantum coherence contributions from the different interaction processes.

IX. CONCLUSIONS

We have presented a fully quantum-mechanical model of the nonlinear interaction of two photons at a two-level atom. The experimental realization of such an interaction can be implemented using a one-sided cavity and single-photon sources. Our theory allows us to determine the effects of an
atomic nonlinearity on the spatiotemporal coherence of a two-photon state. By applying the general results to rectangular input wave packets, we have shown that the nonlinear interaction of two photons at the atom causes bunching and antibunching effects in the two-photon output state. Since our model describes the complete spatiotemporal coherence of the output, it is possible to analyze the origin of these effects in terms of quantum interference between different absorption and propagation processes. This method may therefore provide a useful tool for various applications in the manipulation of individual photons such as quantum information processing, quantum nondemolition measurements, and entangled photon sources.

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