Real-time analysis of two-photon excitation by correlated photons: Pulse-width dependence of excitation efficiency

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We theoretically investigate the dynamics of two-photon excitation by correlated photons with energy anticorrelation in terms of how the excitation efficiency depends on incident pulse width. A three-level atomic system having an intermediate state is used to evaluate the efficiency of two-photon excitation. It is shown that for shorter pulses closer to a monocycle pulse the excitation efficiency by correlated photons is enhanced to become 100 times as large as that by uncorrelated photons.

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I. INTRODUCTION

Two-photon (or multiphoton) excitation is a well known nonlinear optical process and has attracted attention because it has a wide field of application, such as two-photon excitation microscope [1], multiphoton ionization [2], and coherent quantum control [3] in chemical and biochemical fields. The point of two-photon excitation is the coincidence in absorption of photons, and this can be achieved by spatiotemporally controlling light: Light pulsing in the time domain and light focusing in the space domain. Consequently, the photon density interacting with atoms or molecules increases and the efficiency of two-photon excitation is enhanced. In this control method, phase and amplitude play an important role in terms of the degrees of freedom of light.

In a fully quantum-mechanical treatment of light, however, photons exhibit an additional feature, called quantum correlation. Using quantum correlation, we can create a photon pair having inherent coincidence, referred to as squeezed light or quantum-correlated photons. In the field of quantum optics, two-photon excitation by squeezed light has been extensively investigated, and the enhancement of the transition rate has been theoretically predicted [4,5] and experimentally observed [6,7]. A central topic in these early studies is the difference between the intensity dependencies of the transition rate for classical and squeezed light; namely, though the transition rate for classical light has a quadratic dependence on intensity, the transition rate for squeezed light becomes linear at low intensities. Squeezed light is thus essentially different from classical light and behaves as one photon in two-photon excitation at low intensities.

At high intensities, however, the above difference disappears because classical correlation dominates the quantum correlation between the photons constituting squeezed light. In general, the optical nonlinearity requires high intensity owing to its low efficiency. Therefore, if we maximally utilize the quantum nature of light, the efficiency of two-photon excitation at low intensities must be further enhanced. To achieve this, short-pulsed squeezed light at low intensities, ultimately a monocycle pulse composed of two correlated photons, is desired.

In this study, we analyze in detail two-photon excitation exactly by two photons, in terms of how the excitation efficiency depends on incident pulse width. In particular, using a fully quantum-mechanical treatment of light-matter interaction, we investigate the dynamics of two-photon excitation by correlated photons in real time and evaluate the enhancement rate by directly comparing the populations excited by uncorrelated and correlated photons. We show that in the two-photon excitation by correlated photons the simultaneous absorption of two photons effectively takes place and the transition via a virtual state is well suppressed in contrast to classical light input. Furthermore, we show that for shorter pulses close to a monocycle pulse, 100-fold enhancement of excitation efficiency can be realized.

The rest of this paper is organized as follows. In Sec. II, a theoretical model of the atom-cavity system is introduced and the formulation of correlated-photon pairs is given. In Sec. III, we analyze in detail the quantum dynamics of atomic states driven by correlated-photon pairs and the dependence of excitation efficiency on pulse width. In Sec. IV, we summarize and discuss our results.

II. MODEL

A. One-dimensional atom model

As a model system, we consider a one-dimensional photon field interacting with an atom-cavity system, as depicted in Fig. 1(a). An incident two-photon pulse propagates parallel to the r axis and penetrates into a one-sided microcavity (at dumping rate κ). The photons interact with an atom inside the cavity through a cavity field (at coupling rate g) and then turn back to the initial photon field. The atomic system inside the cavity consists of three levels: the ground state $|g\rangle$, the intermediate state $|m\rangle$, and the excited state $|e\rangle$. Their corresponding energies are denoted by ω_m and ω_e (in units of $\hbar$). The central energy of incident photons is set to $k_0 = \omega_e/2$ so that $|e\rangle$ can be resonantly excited. ω_m is far off-resonant with k_0.

In the bad cavity regime of $\kappa \gg g$, photons inside the cavity are emitted so rapidly from the cavity that the cavity field can be adiabatically eliminated. In this case, the atom-photon interaction can be characterized by a single effective emission rate $\Gamma \equiv g^2/\kappa$. If $\Gamma \gg \gamma$, where γ is the spontaneous

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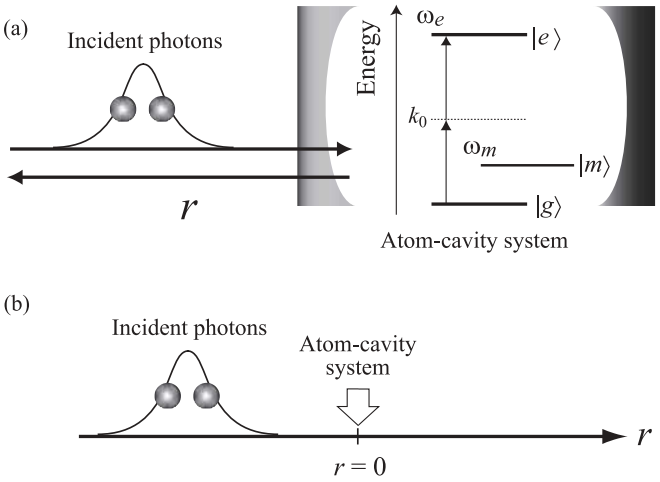

FIG. 1. (a) Schematic of two-photon excitation geometry. (b) One-dimensional atom model.

emission rate of the atomic system into free space (noncavity modes), the system can be reduced to a one-dimensional input-output system with negligible losses [Fig. 1(b)], called the one-dimensional atom model [8]. For simplicity, we ignore nonradiative relaxation losses, such as thermal relaxation.

B. Hamiltonian and quantum dynamics

Setting natural units of $\hbar = c = 1$, the Hamiltonian of the whole system is given by

$$\begin{aligned}\hat{H} = &\int dk\, k\hat{a}_k^\dagger \hat{a}_k + \omega_e|e\rangle\langle e| + \omega_m|m\rangle\langle m| \\ &+ \int dk\sqrt{\Gamma_m/\pi}(|m\rangle\langle g|\hat{a}_k + \hat{a}_k^\dagger|g\rangle\langle m|) \\ &+ \int dk\sqrt{\Gamma_e/\pi}(|e\rangle\langle m|\hat{a}_k + \hat{a}_k^\dagger|m\rangle\langle e|), \qquad (1)\end{aligned}$$

where $\hat{a}_k(\hat{a}_k^\dagger)$ is the annihilation (creation) operator of a photon with energy k. In this study, we ignore direct transition between the states $|g\rangle$ and $|e\rangle$.

The dynamics of the whole system can be calculated from the Schrödinger equation,

$$|\Psi(t)\rangle = \exp(-i\hat{H}t)|\Psi(0)\rangle, \qquad (2)$$

where $|\Psi(0)\rangle$ is the initial state of the whole system, given by

$$|\Psi(0)\rangle = \frac{1}{\sqrt{2}}\int dk \int dk' \psi_{2p}(k,k')\hat{a}_k^\dagger \hat{a}_{k'}^\dagger|0\rangle|g\rangle, \qquad (3)$$

where ψ_{2p} is the two-photon joint amplitude of the incident pulse. The whole wave function is normalized to be $\langle\Psi|\Psi\rangle = 1$. Populations of atomic intermediate and excited states are described by $\langle e\rangle = |\langle e|\Psi(t)\rangle|^2$ and $\langle m\rangle = |\langle m|\Psi(t)\rangle|^2$, respectively.

C. Quantum-correlated photon pair

For comparison, we consider two photon pairs: an uncorrelated photon pair, corresponding to classical light, given by

$$\psi_{2p}(k,k') = \psi(k)\psi(k')e^{-ikr_0}e^{-ik'r_0}, \qquad (4)$$

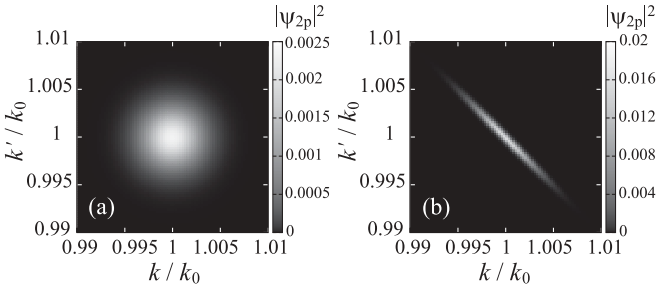

FIG. 2. $|\psi_{2p}|^2$ for (a) uncorrelated and (b) correlated photons. $\sigma \approx 78\lambda$, where $\lambda = 2\pi/k_0$.

and a correlated-photon pair with energy anticorrelation, given by

$$\psi_{2p}(k,k') = \psi(k)\delta(k + k' - \omega_e)e^{-ikr_0}e^{-ik'r_0}, \qquad (5)$$

where $\psi(k)$ is the one-photon pulse and r_0 is the spatial center position of the wave packet at $t = 0$. The correlated photons described by Eq. (5) are referred to as the twin-beam state and can be obtained from spontaneous parametric down-conversion in a beta-barium borate (BBO) crystal [9]. $\delta(k + k' - \omega_e)$ indicates energy anticorrelation of two photons: one photon with energy $k_0 - \Delta k$ is accompanied by another photon with energy $k_0 + \Delta k$, conserving the total energy of $\omega_e = 2k_0$. By Fourier transforming to the time region, this property implies that the photon pair has time coincidence, as has been measured in the famous Hong-Ou-Mandel experiment [10].

According to the spatiotemporal pulse dynamics theory, we define $\psi(k)$ by the Fourier transformation of the space domain and choose $\psi(k)$ having a Gaussian shape,

$$\psi(k)e^{-ikr_0} = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} dr\,\psi(r)\exp(-ikr), \qquad (6)$$

with

$$\psi(r) \propto \exp[-(r - r_0)^2/\sigma^2 + ik_0(r - r_0)], \qquad (7)$$

where σ is the coherent length of the wave packet. The two-photon joint spectra $|\psi_{2p}|^2$ for uncorrelated and correlated photons are shown in Fig. 2. Intriguingly, spectra of the correlated photon pairs are classically identical to those of the uncorrelated photon pair. Thus, the only difference is the quantum correlation, which can be controlled only by quantizing light fields.

III. RESULTS

The dynamics of atomic states induced by quantum-correlated photons can now be calculated by solving Eq. (2). In the actual calculation, we omit the degrees of freedom of polarization by assuming linearly polarized light and numerically solve Eq. (2) by discretizing the photon fields. Further, we approximate the δ function in Eq. (5) by a Gaussian function having a width $\sigma/8$, corresponding to a typical value of correlated photons obtained from a BBO crystal. As a model of the atomic system, we adopt energies of a Cs atom and choose the energy levels of $\omega_{e_A} = 2k_0$ and $\omega_{m_A} = 0.86k_0$ for $|e\rangle$ and $|m\rangle$, respectively. For simplicity, we choose the same value of $\Gamma_e = \Gamma_m \approx 0.003k_0$ in each level.

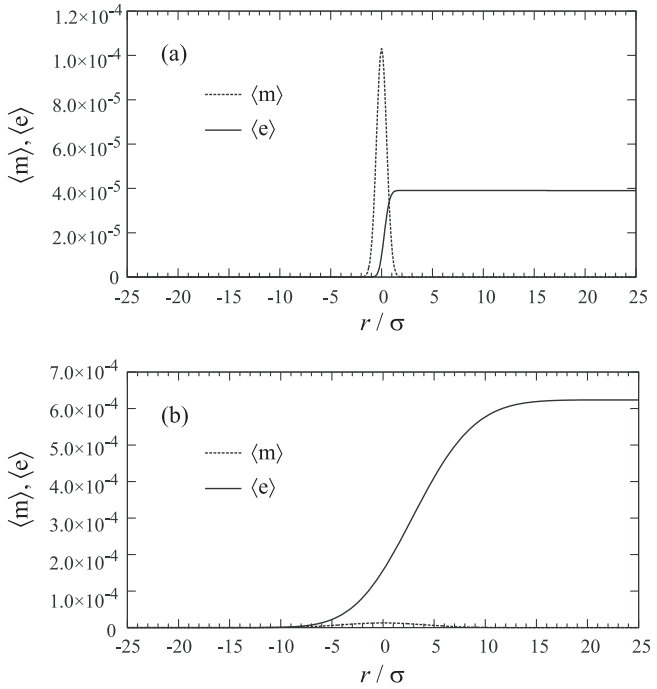

FIG. 3. $\langle e\rangle$ and $\langle m\rangle$ as a function of r for (a) uncorrelated photons and (b) correlated photons.

Figure 3 shows the population dynamics induced by uncorrelated and correlated photons. The parameters are the same as those in Fig. 2. The horizontal axis is the spatial center position r of the incident pulse, normalized by σ, and the vertical axis is the population of the atomic system, where the solid and dashed curves represent $\langle e\rangle$ and $\langle m\rangle$, respectively. For uncorrelated photons (a), when an incident pulse reaches $r = 0$, the atomic system absorbs one photon and $\langle m\rangle$ is excited first. Then $\langle e\rangle$ is excited by absorbing another photon and $\langle m\rangle$ disappears. This process is the well known two-photon excitation via a virtual state. The efficiency of two-photon excitation becomes large only when the subsequent photon comes to the atomic system while the virtual state survives. Therefore, photon coincidence is crucial in the two-photon excitation. For correlated photons (b), however, the virtual state $\langle m\rangle$ is hardly excited. Instead, $\langle e\rangle$ absorbs two photons simultaneously and is drastically enhanced. In the present parameters, the enhancement of $\langle e\rangle$ is about 15-fold greater compared to uncorrelated photons. Thus, owing to the coincidence of photons originating from quantum correlation, the enhancement of two-photon excitation is achieved.

Of particular interest is that simultaneous absorption of two photons effectively occurs despite no interaction terms such as $|e\rangle\langle g|\hat{a}_k\hat{a}_{k'} + \hat{a}_k^\dagger\hat{a}_{k'}^\dagger|g\rangle\langle e|$ in Eq. (1). This is because photons constituting a correlated-photon pair are inseparable as one quantum state. As can be seen in Eq. (5), correlated photons cannot be divided into the product of one-photon states as in Eq. (4), and hence excitation of $\langle m\rangle$ hardly takes place. As a result, efficient two-photon excitation is achieved.

Figure 4 shows the dependence of enhancement β on σ normalized by the central wavelength λ of the incident pulse. The parameters are the same as those in Fig. 2 except σ. β is defined by the ratio of $\langle e\rangle_{\text{corr.}}/\langle e\rangle_{\text{uncorr.}}$, where $\langle e\rangle_{\text{corr.}}$ and $\langle e\rangle_{\text{uncorr.}}$ are the populations excited by correlated and uncorrelated photons, respectively, and are obtained after an incident photon pulse completely passes through an atomic system. In the excitation exactly by two photons, the efficiency by correlated photons always dominates that by uncorrelated photons. For large σ (long pulse), β is small because of little quantum correlation. As σ increases, however, β increases owing to the increase of coincidence of two photons. In the present parameters, β reaches $\sim$100. Although we only calculate up to $\sigma/\lambda \approx 10$ because of the difficulty in the calculation due to discretization of the photon field, β will increase for further decreases in σ. In particular, for a monocycle pulse ($\sigma/\lambda \approx 1$), large β can be expected so that efficient two-photon excitation could be achieved by one photon pair.

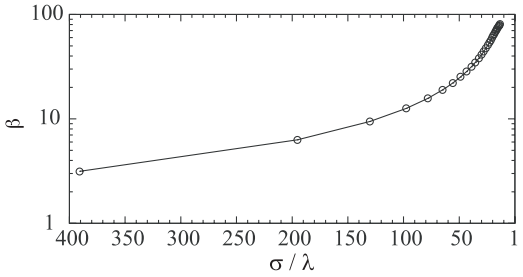

FIG. 4. β as a function of σ.

IV. CONCLUSION AND DISCUSSION

We have theoretically investigated the dynamics of two-photon excitation by correlated photons using a fully quantum-mechanical treatment, in terms of the dependence of excitation efficiency on incident pulse width. We have shown that in the excitation exactly by two photons the excitation efficiency by correlated photons is always enhanced compared to uncorrelated photons and the enhancement rate becomes more prominent for shorter pulses.

Throughout this work, we have barely mentioned the generating methods of correlated photons. Here we discuss two candidates of real systems, as examples, in which required photon pairs with small σ could be achieved. One is parametric down-conversion utilizing the quasiphase matching condition (QPMC). In general, in nonlinear crystals, such as a BBO crystal, σ is noncontrollable. However, a QPMC system can control the two-photon state by artificially designing the material. In fact, correlated photons with small $\sigma \sim 10$ fs have already been reported [11,12]. Another is to utilize cavity quantum electrodynamics (QED) effects realizable in an exciton system confined in a semiconductor microcavity. The QED system can also control the two-photon state by exciton-cavity designing, and correlated photons can be efficiently generated (e.g., by utilizing a biexciton state [13–15]).

Finally, we discuss a possibility of further enhancement of excitation efficiency and a correlated photon pair with time delay. In this study, we have replaced the δ function in Eq. (5) by the Gaussian function. By squeezing the Gaussian shape closer to the δ function (to smaller width), enhancement of efficiency and highly selective excitation can be achieved [16]. If the present results are combined with the method in Ref. [16], a huge enhancement of excitation efficiency more

than 1000 times can be expected. Further, by introducing a time delay between two photons, the calculation method presented in this work can be directly applied to an analysis of up-conversion by entangled photons as reported in Ref. [17].

We hope that the results of this study facilitate applications of correlated photons to optical devices based on two-photon excitation.

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