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Generation of photon pairs using polarization-dependent two-photon absorption

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We propose a method for generating photon pairs from coherent light, using polarization-dependent two-photon absorption. We study the photon statistics of two orthogonally polarized modes by solving a master equation, and show that when we prepare a coherent state in one polarization mode, photon pairs are created in the other mode. The photon pairs have the same frequency as that of the incident light.

I. INTRODUCTION

A photon pair shows a variety of nonclassical phenomena such as nonlocal quantum correlation [1], two-photon interference [2], and so on. There are interesting applications that include quantum teleportation [3], quantum cryptography [4], and quantum computing [5]. Usually photon pairs are generated with two-photon cascade emission [6] or spontaneous parametric down-conversion [7,8]. In this paper we propose a method to produce photon pairs, using polarization-dependent two-photon absorption from coherent light.

Two-photon absorption is one of the second-order nonlinear optical effects and many investigations have been performed [9,10]. Some researchers have shown that coherent light is changed into sub-Poissonian light with nonclassical characteristics after the interaction with the two-photon absorbers [11–13]. The rate of two-photon transition is proportional to the second-order correlation function of the photon field [14]. The shorter the time interval between two photons incident on a two-photon absorber, the higher the probability of the absorption. If the initial state of the field is a coherent state whose photons are randomly distributed in time, pairs of photons that arrive at the absorber accidentally within a very short time period are likely to be absorbed. Hence the unabsorbed photons are apart from each other, and exhibit antibunched photon statistics with low second-order coherence [15].

The above investigations deal with the polarization-independent two-photon transition. But there exists two-photon transition with a polarization selection rule that allows only the transition by two photons whose polarizations are orthogonal to each other. When a polarization-dependent two-photon absorber interacts with a coherent light that includes quantum superposition of an absorbable and an unabsorbable two-photon states, the time-correlated photon pairs in the unabsorbable state and isolated photons survive after the absorption. The polarization selective absorption accompanies a change of the polarization state for the photon pairs, and therefore, photon pairs with polarization orthogonal to the original polarization are created. One can single out the photon pairs using a linear polarizer. We will give the details in Sec. II and analyze the time evolution of the field in two orthogonal linear polarization modes, using a master equation in Sec. III.

While photon pairs created by parametric down-conversion have half the frequency of the pump laser, the frequency of photon pairs generated by our method is the same as that of the coherent light initially prepared. Hence, choosing appropriate two-photon transitions, one could produce photon pairs in blue or much higher frequency region.

II. POLARIZATION-DEPENDENT TWO-PHOTON ABSORPTION

The two-photon transition between two $L=0$ states obeys a polarization selection rule that only allows the absorption of oppositely circular-polarized photons [16,17]. The selection rule is originated from the conservation of angular momentum of the atom and the two photons, i.e., the photons must have zero angular momentum if the initial state and the final state of the atom have zero angular momentum. If one photon is right circularly polarized, the other must be left circularly polarized.

In this section, we will give simple explanations how photon pairs are generated by the polarization-dependent two-photon absorption. We introduce the two-photon state $|\Psi_a\rangle = |1,1\rangle_{RL}$, where $|r,l\rangle_{RL}$ denotes the state with r right circularly polarized photons and l left circularly polarized photons. We can represent $|\Psi_a\rangle$ in the linear polarization basis as

$$|\Psi_a\rangle = \frac{|2,0\rangle_{HV} + |0,2\rangle_{HV}}{\sqrt{2}}, \qquad (1)$$

where $|h,v\rangle_{HV}$ is the state with h photons in the horizontal polarization mode and v photons in the vertical polarization mode [18].

First, we consider a simple case where photon field is prepared in $|\Psi_i\rangle = |2,0\rangle_{HV}$ and interacts with the polarization-dependent two-photon absorber. If the interaction time is long enough, the state of the light is projected into the state which is orthogonal to the state $|\Psi_a\rangle$, as illustrated in Fig. 1(a). Then the conditioned state for no absorption is represented by

$$|\Psi_f\rangle = (1 - |\Psi_a\rangle\langle\Psi_a|)|2,0\rangle_{HV} = \frac{|2,0\rangle_{HV} - |0,2\rangle_{HV}}{2}, \qquad (2)$$

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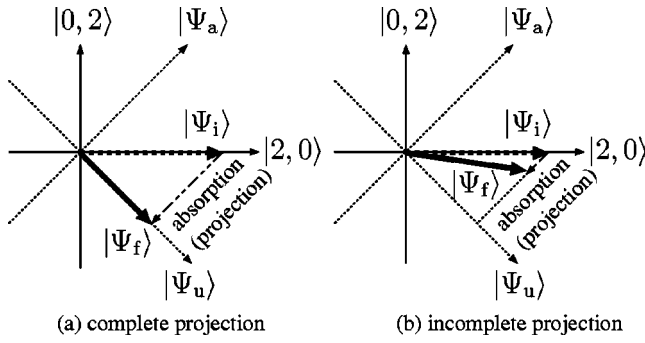(a) complete projection   (b) incomplete projection

FIG. 1. Projection of the state by polarization-dependent two-photon absorption. The unabsorbable state $|\Psi_u\rangle$ is defined as $(|2,0\rangle - |0,2\rangle)/\sqrt{2}$.

where the decrease of the norm is the result of the absorption. Note that there is a probability that one finds the vertically polarized two-photon state $|0,2\rangle_{HV}$ that does not exist before the interaction. A similar process for one photon is occurred when a horizontally polarized photon passes through a linear polarizer aligned at 45°. The polarization is projected along the orientation of the polarizer, and the vertical polarization component appears [19].

In the case of incomplete absorption, as shown in Fig. 1(b), the conditioned state for no absorption is given by

$$|\Psi_f\rangle = (1 - \epsilon|\Psi_a\rangle\langle\Psi_a|)|2,0\rangle_{HV}$$

$$= \left(1 - \frac{\epsilon}{2}\right)|2,0\rangle_{HV} - \frac{\epsilon}{2}|0,2\rangle_{HV}, \quad (3)$$

where ϵ (>0) represents absorption. For $\epsilon \ll 1$, the probability for $|2,0\rangle_{HV}$ is reduced by ϵ and that for $|0,2\rangle_{HV}$ is increased by $\epsilon^2/4$. This change in polarization creates the vertically polarized two-photon state, which did not exist initially.

Next, we consider the case where the initial state of light is a horizontally polarized coherent state as shown in Fig. 2. Because the rate of the two-photon absorption is proportional to the second-order correlation function of the field, pairs within an extremely short interval are absorbed selectively from the randomly distributed photons. Therefore, only the time-correlated pairs are changed into the superposition as Eq. (3) by the absorption. The pairs with horizontal polarization are reduced, while vertically polarized photon pairs emerge with probability $\epsilon^2/4$ for all pairs. All uncorrelated and isolated photons remain horizontally polarized.
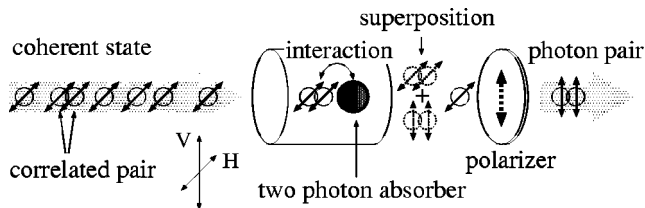

FIG. 2. Procedure of generating photon pairs with a two-photon absorber.

The horizontally polarized beam is evolved from the coherent state into antibunched light due to the pairwise absorption. On the other hand, the vertical polarization mode is a vacuum at first, and evolved into the bunching state by pair production of photons. As shown in Fig. 2, filtering out horizontally polarized photons with a polarizer, one can get photon pairs.

In the following section, we will analyze the photon statistics of the horizontal and vertical polarization mode for quantitative evaluations.

III. MASTER EQUATION APPROACH

We examine the time evolution of the field under the interaction with the polarization-dependent two-photon absorbers by solving a master equation. Schauer [20] investigated the steady state solutions of the master equation for the interaction between the two-photon absorbers and the photon field. On the other hand, we take a perturbative approach under the condition of short interaction time.

We assume that the one-photon transitions to intermediate levels are off resonant enough to be negligible compared with the two-photon transition. We introduce a two-photon annihilation operator O:

$$O = a_R a_L = \frac{a_H^2 + a_V^2}{2}, \quad (4)$$

where a_R, a_L, a_H, and a_V are annihilation operators for the right circularly, left circularly, horizontally, and vertically polarized photons, respectively. The density matrix of the light field, ρ, satisfies the following master equation [12,20]:

$$\frac{d\rho}{d\tau} = 2O\rho O^\dagger - O^\dagger O\rho - \rho O^\dagger O, \quad (5)$$

where the time τ is normalized with respect to the two-photon transition rate κ as $\tau = \kappa t$. Equation (5) yields the equation of motion for an operator A as

$$\frac{d}{d\tau}\langle A\rangle = 2\langle O^\dagger A O\rangle - \langle A O^\dagger O\rangle - \langle O^\dagger O A\rangle. \quad (6)$$

We will calculate the evolution of the light field when the initial state of the light is an intense coherent state $|\alpha\rangle_H$ for the horizontal polarization mode and the vacuum state $|0\rangle_V$ for the vertical polarization mode, i.e., $|\alpha,0\rangle_{HV} \equiv \exp(\alpha a_H^\dagger - \alpha^* a_H)|0\rangle$. As mentioned before, we assume that the normalized interaction time T is much less than unity. This is reasonable for general experimental realizations without special equipments such as a high-Q cavity because of very small two-photon transition rate.

A. Horizontal polarization mode

With Eq. (6), the rate of change for the number of horizontally polarized photons can be easily obtained as

$$\frac{d}{d\tau}\langle a_H^\dagger a_H\rangle = -\langle a_H^{\dagger 2} a_H^2\rangle - \frac{1}{2}\langle a_H^{\dagger 2} a_V^2\rangle - \frac{1}{2}\langle a_H^2 a_V^{\dagger 2}\rangle. \quad (7)$$
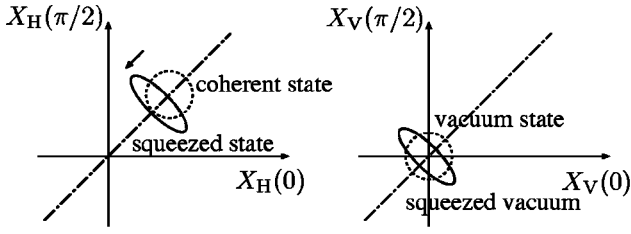
(a) horizontal polarization mode (b) vertical polarization mode

FIG. 3. Time evolution in the phase space (a) for the horizontal polarization mode and (b) for the vertical polarization mode. The dotted lines represent initial states and the solid lines represent the state after the interaction with the absorber.

The interaction time is so small that the horizontally polarized beam still remains to be very intense and the vertical polarization is nearly in the vacuum mode after the interaction. Then, the first term of the right-hand side of Eq. (7) is dominant:

$$\frac{d}{d\tau}\langle a_H^\dagger a_H\rangle \simeq -\langle a_H^{\dagger 2} a_H^2\rangle. \tag{8}$$

The reduction rate of the photon number is proportional to the second-order correlation function of the field.

From Eq. (8), the mean photon number after the interaction can be approximated as

$$\langle a_H^\dagger a_H\rangle \simeq |\alpha|^2 - |\alpha|^4 T. \tag{9}$$

Similarly, other resulting expectation values are calculated as

$$\langle a_H\rangle \simeq \alpha - \frac{|\alpha|^2\alpha T}{2},$$

$$\langle a_H^2\rangle \simeq \alpha^2 - |\alpha|^2\alpha^2 T - \frac{\alpha^2 T}{2}, \tag{10}$$

$$\langle a_H^{\dagger 2} a_H^2\rangle \simeq |\alpha|^4 - 2|\alpha|^6 T - |\alpha|^4 T.$$

We introduce a Hermitian operator $X_H(\phi) = a_H e^{-i\phi} + a_H^\dagger e^{i\phi}$, and calculate its dispersion $\langle[\Delta X_H(\phi)]^2\rangle$ from Eqs. (9) and (10):

$$\langle[\Delta X_H(\phi)]^2\rangle \simeq 1 - |\alpha|^2 T\cos 2(\arg\alpha - \phi), \tag{11}$$

which shows that the dispersions of the quadrature amplitudes $X_H(\arg\alpha)$ and $X_H(\arg\alpha + \pi/2)$ satisfy $\langle[\Delta X_H(\arg\alpha)]^2\rangle < 1 < \langle[\Delta X_H(\arg\alpha + \pi/2)]^2\rangle$. We see that the mode of the horizontal polarization is evolved into the squeezed state, where the dispersion for $\phi = \arg\alpha$ is smaller than unity. As in Fig. 3(a), the fluctuation in the direction of the excitation of the coherent state decreases and the photon statistics become sub-Poissonian.

B. Vertical polarization mode

We will calculate the photon number in the vertical polarization mode, $\langle a_V^\dagger a_V\rangle$, and the second-order correlation function $\langle a_V^{\dagger 2} a_V^2\rangle$. They are approximated as

$$\langle a_V^\dagger a_V\rangle = \langle a_V^{\dagger 2} a_V^2\rangle = \frac{(|\alpha|^2 T)^2}{4}, \tag{12}$$

which shows that the number of the vertically polarized photons, as well as the second-order correlation function, grows in proportion to the second-order correlation function of the horizontal polarization mode. The normalized second-order correlation function is obtained as

$$g^{(2)}(0) = \frac{\langle a_V^{\dagger 2} a_V^2\rangle}{\langle a_V^\dagger a_V\rangle^2} = \frac{4}{(|\alpha|^2 T)^2} \gg 1. \tag{13}$$

Owing to the pair creation of photons, the strongly bunched photons appear in the vertical polarization mode.

Similar calculations lead to $\langle a_V\rangle = 0$ and $\langle a_V^2\rangle = -\alpha^2 T/2$. The evolution of $\langle a_V^2\rangle$ is sensitive to the phase of the coherence of the horizontal polarization mode. The dispersion $\langle[\Delta X_V(\phi)]^2\rangle$ of an Hermitian operator $X_V(\phi) = a_V e^{-i\phi} + a_V^\dagger e^{i\phi}$ is approximated as

$$\langle[\Delta X_V(\phi)]^2\rangle = 1 - |\alpha|^2 T\cos 2(\arg\alpha - \phi), \tag{14}$$

which is same as Eq. (11). The state of the vertically polarized photons is determined by that of the horizontally polarized photons and the directions of squeezing are same. Figure 3(b) illustrates the evolution from the vacuum state to the squeezed vacuum in the phase space. Unlike the coherent squeezed state in Fig. 3(a), the squeezed vacuum exhibits super-Poissonian statistics [21].

IV. CONCLUSION AND DISCUSSION

We have proposed the unique method of generating photon pairs with the polarization-dependent two-photon absorption. The results of the perturbative calculations with the master equation show that the vertically polarized photons and its second-order correlation function are increased together with the second-order correlation function of the horizontally polarization mode. Moreover, the phase space representation shows that the coherent state in the horizontal polarization mode is changed into antibunching state, and the vacuum state in the vertical polarization mode is developed into a squeezed vacuum whose photon statistics is bunched. The photon bunching is caused by the pair creation.

The generation of photon pairs at frequency ω_0 by parametric down-conversion requires a pump beam at frequency $2\omega_0$, so it is difficult to obtain photon pairs in the blue or shorter wavelength region. On the other hand, all photons in our method have the same frequency ω_0. By using appropriate two-photon absorbers, we could produce photon pairs in blue or higher frequency range.

We estimate the possibility of experimental implementation when we use a cw laser. Using the two-photon absorption coefficient β, κ introduced in Sec. III is expressed as

$\kappa = \beta \hbar \omega_0 c/(n^2 AL)$, where n, A, and L are the refraction index, the laser beam cross section, and the interaction length, respectively. For the 6S-8S two-photon transitions of cesium at 822 nm, the two-photon absorption coefficient is $\beta \sim 1.8 \times 10^{-11}$ m/W for atomic vapor at room temperatures. When setting $L \sim 1 \times 10^{-2}$ m and the laser intensity $I \sim 1 \times 10^{10}$ W/m^2, and assuming $n \sim 1$, one obtains $N_{\mathrm{pair}} = c\beta^2 I^2 L/(4n) \sim 2.4 \times 10^4$ photon pairs per second. Although N_{pair} seems to be a moderate value, the experiment under this condition may be hard to carry out. It is difficult to separate photon pairs from intense laser beam with orthogonal polarization. For the laser beam cross section $A \sim 10^{-10}$ m^2, the ratio between N_{pair} and the photon number of the laser beam per second $N_{\mathrm{laser}} = AI/(\hbar \omega_0)$ is given as $R \equiv N_{\mathrm{pair}}/N_{\mathrm{laser}} \sim 5.6 \times 10^{-15}$, but the extinction ratio of commercially available linear polarizers is 10^{-6} at best. The ratio R can be improved by increasing the atomic density or confining the vapor in a cavity.

It is known that the two-photon transition for the Γ_1 biexciton in CuCl has the same selection rule [22]. It has the advantage of large two-photon absorption coefficient $\beta \sim 10^{-3}$ m/W. Other parameters are $n \sim 3$ and $\hbar \omega_0 = 3.2$ eV. If we choose $I \sim 10^4$ W/m^2 and set the other parameters the same as in the case of Cs, we get $N_{\mathrm{pair}} \sim 2.5 \times 10^7$/s and $R \sim 1.2 \times 10^{-5}$, which is large enough to discriminate. While we have assumed the use of cw lasers for estimation, the use of a pulsed laser would certainly ease the experimental conditions.

In this paper we have dealt with the degenerate two-photon transition, but it is easy to extend our method to a nondegenerate transition. In the degenerate two-photon process, two photons appear simultaneously in pair in the same mode, while in the nondegenerate process the pair of photons will emerge simultaneously but in different frequency modes. In the nondegenerate case, it is possible to adjust the detuning Δ from the intermediate level and we can easily attain large two-photon absorption coefficient β, which is inversely proportional to Δ^2. This strategy might enable us to demonstrate the experiments using atomic vapor.

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