Generation of Phase States by Two-Photon Absorption

Hiromi Ezaki
Faculty of Engineering, Tokyo Institute of Polytechnics, 1583 Iiyama, Atsugi-shi, Kanagawa 243-0297, Japan

Eiichi Hanamura
Optical Sciences Center, The University of Arizona, Tucson, Arizona 85721

Yoshihisa Yamamoto
Quantum Entanglement Project, ICORP, JST, E.L. Ginzton Laboratory, Stanford University, Stanford, California 94305-4085
(Received 2 September 1998)

A new method for producing a phase state by two-photon absorption is proposed. We show that such a process conserves the phase of an initial coherent state $|\alpha\rangle$ and converts it to $|\psi\rangle = (|0\rangle + e^{i\phi}|1\rangle)/\sqrt{2}$, where $\alpha = |\alpha|e^{i\phi}$. Therefore, we obtain desirable phase states by controlling the phase of the initial coherent state. Appropriate materials with a reasonable two-photon absorption rate are proposed.

PACS numbers: 03.65.Bz, 42.50.Dv, 42.50.Lc

Generation and control of a single-photon state and a quantum bit (qubit) state are extensively studied from both viewpoints of fundamental interest and applications to quantum cryptography and communication. A popular method for producing a single-photon state is to use one of the twin photons created by parametric down-conversion as a signal and the other as a trigger [1]. The generation time of a photon pair is randomly distributed according to the Poisson-point process. A single-photon turnstile device, based on simultaneous Coulomb blockade for electrons and holes in a mesoscopic *pn* junction, was proposed [2] and demonstrated [3] to generate heralded single photons. The repetition rate is limited by a radiative recombination lifetime. Recently, Imamoglu and colleagues [4] proposed "photon blockade," in which optical tunneling is prevented by Kerr nonlinearity instead. They avoid the usual one-photon absorption using atomic dark resonance. To avoid one-photon loss, a special condition must be satisfied for the pulse intensity and width, which places also a limit on the repetition rate.

A 50%–50% beam splitter converts a single-photon state into a quantum entangled state, $(|10\rangle + e^{i\phi}|01\rangle)/\sqrt{2}$. The two outputs of the beam splitter are correlated, so that, if one of the two beams is lost to reservoirs, the other is collapsed to either $|0\rangle$ or $|1\rangle$. Generation of a single qubit state $(|0\rangle + e^{i\phi}|1\rangle)/\sqrt{2}$, which is proposed in this paper, is preferred for some applications because there is no need to protect the correlated counterpart.

In this paper, we propose a new method for generating phase states $(|0\rangle + e^{i\phi}|1\rangle)/\sqrt{2}$, which is the special case of a Pegg-Barnett phase state [5], by two-photon absorption (TPA). The present phase state is considered as a qubit state in the language of quantum information science. Let us consider a series of well-focused short laser pulses to be irradiated on a side of crystal in which TPA is effective. Under these conditions, the radiation field is well confined to a small volume. Therefore we may expect the TPA rate becomes larger than a single-photon one even when a laser pulse contains two or three photons. We will discuss several possible systems, i.e., atomic gas, exciton-biexciton system, and semiconductors. We choose, e.g., for the last case, the fundamental frequency ω well below the exciton frequency, but the two-photon frequency 2ω is well above the band-to-band transition frequency. The laser pulses are attenuated dominantly by TPA and each becomes a superposition of single- and zero-photon states with almost equal amplitude.

A similar qubit state can be generated by the interaction between a single mode high-Q cavity field and a two-level atom [6,7], but the extraction of such a state out of the cavity with a high repetition rate for measurement and communication is nontrivial.

We have chosen the carrier frequency enough off resonant from the intermediate levels. Then, attenuation rate κ_1 due to single-photon absorption becomes smaller than electronic excitation rate κ_2 by two-photon transition. The electronic state excited by TPA is embedded in continuum states so that its relaxation rate is large enough. As a result, both the intermediate and final electronic states in the total density matrices $\rho^{\mathrm{tot}}(\tau)$ can be adiabatically eliminated.

When we introduce time coordinate $t = \tau - z/v_g$ moving with the laser pulse peak with group velocity v_g in the z direction, the master equation of the radiation field is derived as follows [8,9]:

$$\frac{\partial \rho}{\partial t} = -\kappa_1(b^\dagger b\rho - 2b\rho b^\dagger + \rho b^\dagger b) - \kappa_2(b^{\dagger 2}b^2\rho - 2b^2\rho b^{\dagger 2} + \rho b^{\dagger 2}b^2), \quad (1)$$

where $b^\dagger$ and b are creation and annihilation operators of photons composing a light pulse, κ_1 is the rate of a one-photon absorption, and κ_2 is that of the TPA. These rates κ_1 and κ_2 are related to a single- and two-photon

 0031-9007/99/83(17)/3558(4)$15.00

absorption coefficients $\alpha^{(1)}$ and $\alpha^{(2)}$ by $\alpha^{(1)} = \kappa_1/v_g$ and $\alpha^{(2)} = \kappa_2 n/v_g$ with photon number n. The master equation is based on an effective Hamiltonian that ignores the dynamical Stark shift [10], which brings the constant energy shift of the field and the photon-number-dependent energy shift of the atomic system. The latter term is proportional to the difference between the squares of the coupling constants with the field g_1 (for $|g\rangle \rightarrow |i\rangle$) and g_2 (for $|i\rangle \rightarrow |e\rangle$), where g, i, and e are the electronic ground, the intermediate, and the excited states. Therefore, if $g_1 \sim g_2$, the latter term becomes negligible compared to the two-photon Rabi frequency, which is also negligible compared to the relaxation rate of the excited state. Then only the carrier frequency is modified by constant. This is applied to the exciton-biexciton system and the atomic system we consider in this paper.

First, we consider an ideal two-photon absorber; i.e., κ_1 is negligibly small compared with κ_2. Then the steady state solution of Eq. (1) is analytically solved:

$$\langle 0|\rho|0\rangle = \sum_{q=0}^{\infty} \langle 2q|\rho(0)|2q\rangle, \tag{2}$$

$$\langle 1|\rho|1\rangle = \sum_{q=0}^{\infty} \langle 2q+1|\rho(0)|2q+1\rangle, \tag{3}$$

$$\langle 0|\rho|1\rangle = \langle 0|\rho(0)|1\rangle + \sum_{q=1}^{\infty} \prod_{r=1}^{q} \sqrt{1 - \frac{1}{4r^2}} \langle 2q|\rho(0)|2q+1\rangle. \tag{4}$$

When the initial state is a highly excited coherent state $|\psi_{\rm in}\rangle = |\alpha\rangle$ ($\alpha = |\alpha|e^{i\phi}, |\alpha| \gg 1$), we obtain the output state $|\psi_{\rm out}\rangle$:

$$|\psi_{\rm out}\rangle = \frac{1}{\sqrt{2}} (|0\rangle + e^{i\phi}|1\rangle), \tag{5}$$

which is numerically confirmed. Therefore, we obtain phase states (qubit state with arbitrary phase) by controlling the phase of an initial coherent state. The initial phase is conserved through a two-photon damping process. This marked property is the characteristic of the TPA process.

The mechanism of the phase conservation will be understood as follows: First, in TPA, the damping process is divided into two channels: even and odd photon number. Suppose the initial coherent state is expanded by the Fock states:

$$|\alpha\rangle = \sum_{n=0}^{\infty} c_n|n\rangle. \tag{6}$$

The phase difference between c_{2n+1} and c_{2n} equals ϕ: $c_{2n+1}/c_{2n} \sim |\alpha|e^{i\phi}$. Second, when one TPA event occurs, the wave function changes $\sum c_n|n\rangle \rightarrow \sum c_n\sqrt{n(n-1)}|n-2\rangle$ except for the normalization constant. Therefore, the phase of the wave function does not change in this transition. As a result, the n-odd and n-even channels keep the phase difference in TPA, so that the initial coherent state is converted to the phase state [Eq. (5)] except for the absolute phase factor due to free evolution.

The dynamics of the wave function is clearly demonstrated by a quantum Monte Carlo wave function simulation, or quantum trajectory method [11]. Figure 1 illustrates a single trajectory showing the time evolution of the expansion coefficients c_n for the initial coherent state $|\alpha\rangle$, where $\alpha = -5$. We clearly observe that the c_n with even n take positive values, whereas the c_n with odd n take negative values. The phase difference π is conserved and the final state approximately approaches to $(|0\rangle - |1\rangle)/\sqrt{2}$. Figure 2 shows the steady state expansion coefficients averaged over 1000 trajectories. We observe that a phase state [Eq. (5)] is obtained for $\alpha > 5$. We also observe from Fig. 2 that, if we start from a coherent state with small average photon number, we obtain qubit states with different probability amplitudes,

$$c_0|0\rangle + c_1 e^{i\phi}|1\rangle, \tag{7}$$

with $c_0/c_1 > 1$, which covers the lower hemisphere of the entire Bloch sphere.

Next, we consider the effect of the one-photon absorption which is not negligible in realistic systems. However, if $\kappa_2 > \kappa_1$, the state above is transiently realized. Figure 3 shows the density matrix elements ρ_{00}, ρ_{11}, and ρ_{01} as a function of time obtained by solving Eq. (1) numerically. When the linear damping rate is much weaker than the nonlinear one, e.g., $\kappa_2/\kappa_1 = 10$, one- and zero-photon states can be obtained with almost the same probabilities. When we set the length ($l_s \equiv v_g t_s$) of the material corresponding to $\kappa_2 t_s = 0.6$ denoted by the arrow in Fig. 3, we obtain the above-mentioned phase state with high efficiency. For example, an adequate length is $l_s = 4 \sim 7$ mm for $\kappa_2/\kappa_1 = 10$ when we choose $t_s = 0.4 \sim 0.7 \times 10^{-10}$ s and $v_g \sim 10^9$ cm/s. The efficiency of the generation of a

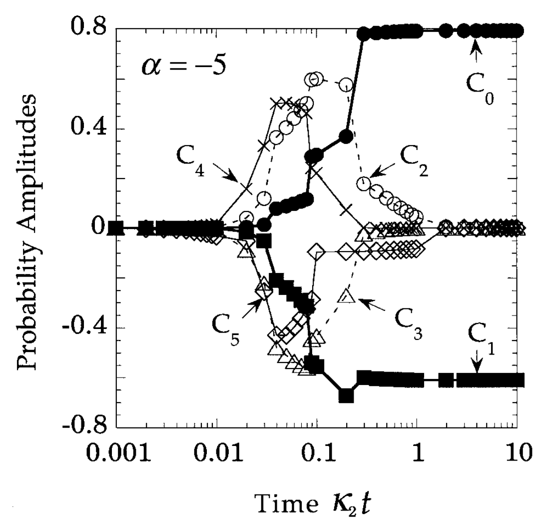

FIG. 1. A single quantum trajectory showing the probability amplitudes c_n as a function of time. The initial state is a coherent state $|\alpha = -5\rangle$.

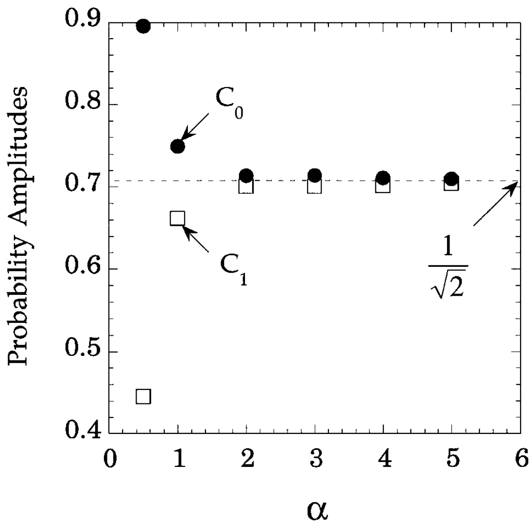

FIG. 2. Probability amplitudes c_0 and c_1 averaged over 1000 trajectories as a function of α.

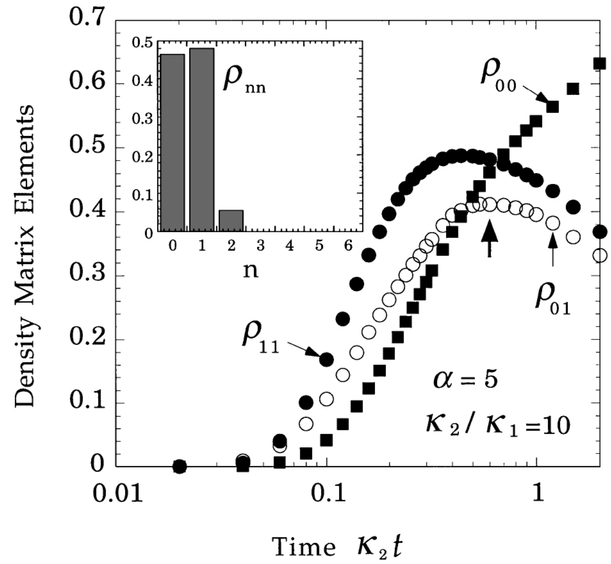

FIG. 3. Density matrix elements, ρ_{00}, ρ_{11}, and ρ_{01} as a function of time. Inset shows the distribution of the photon number at the optimum interaction time $\kappa_2 t = 0.6$.

phase state depends on the ratio κ_2/κ_1 as shown in Fig. 4. We plot ρ_{00}, ρ_{11}, and ρ_{01} at the optimum time when the off-diagonal element ρ_{01} takes a maximum value. To obtain a phase state with high efficiency, $\kappa_2/\kappa_1 > 10$ should be satisfied, which is realizable as will be discussed later.

Before proceeding, we show the self-phase modulation to be negligible. TPA occurs stochastically in time, which seems to dephase the quantum coherence between $|0\rangle$ and $|1\rangle$ states via the photon-number-dependent phase rotation by the Hamiltonian $H_0 = \hbar\omega b^\dagger b + \hbar\chi\hat{n}(\hat{n}+1)$ for free evolution (first term) and self-phase modulation (second term). However, we can prove that this dephasing effect is negligible by the following simple argument. Suppose the initial coherent state is expanded by the Fock states as in Eq. (6). Then each Fock state $|n\rangle$ evolves with the effective Hamiltonian $H = H_0 + V$. Here, $V = V^{(2)}bb + \text{H.c.}$, where the coordinates of the electronic system are taken into account in the coefficient $V^{(2)}$ through the TPA coefficient κ_2 by $V^{(2)} = i\kappa_2$. Here and hereafter, we put $\hbar = 1$. Then the time development is expanded in V as follows:

$$e^{-iHt}|n\rangle = \left\{ e^{-iH_0 t} - i\int_0^t dt_1\, e^{-iH_0(t-t_1)} V e^{-iH_0 t_1} + (-i)^2 \int_0^t dt_1 \int_0^{t_1} dt_2\, e^{-iH_0(t-t_1)} V e^{-iH_0(t_1-t_2)} V e^{-iH_0 t_2} + \cdots \right\} |n\rangle . \tag{8}$$

The first, second, and third terms of the right-hand side of Eq. (8) correspond to the cases with no TPA, one TPA at t_1, and two TPA at t_2 and at t_1, in this order. The self-phase modulation for the single TPA event at $t = t_i$ is different from the one for no TPA event by $e^{i\chi(n_i-1)(n_i-2)t_i} e^{-i\chi n_i(n_i+1)t_i} = e^{-i\chi(4n_i-2)t_i}$, where n_i is the photon number just before t_i. The average time uncertainty Δt_i in which single TPA for n_i is induced is $\Delta t_i \sim 1/[\kappa_2 n_i(n_i - 1)]$. As a result, the phase uncertainty associated with a random single TPA event is evaluated as $\chi(4n_i - 2)/[\kappa_2 n_i(n_i - 1)]$ for $n_i \geq 2$. After such self-phase modulation is accumulated for an order of $(n_i/2)$ events, total phase uncertainty is an order of $2\chi/\kappa_2$. Remember that $\chi \sim N|P_{gi}|^4/[\hbar^3(\omega - \omega_{ig})^3]$, and $\kappa_2 \sim N|P_{gi}|^2|P_{ie}|^2/[\hbar^3(\omega - \omega_{ig})^2\Gamma_2]$, where Γ_2 is the relaxation rate of the excited state, and N is a number density of oscillators times $(e/m)^4[\hbar/(2\epsilon_0 V\omega)]^2$. Thus the total phase uncertainty $\chi/\kappa_2 \sim \Gamma_2/|\omega - \omega_{ig}|$. As long as a single-photon energy is enough off resonant from the first excitation $\hbar\omega_{ig} = E_i - E_g$, which is our case, the self-phase modulation effect is negligible. As a result, the phase randomization between $|0\rangle$ and $|1\rangle$ states are shown to be negligible against the probability distribution of TPA events $(t_1, t_2, \ldots, t_{n/2})$.

Next, we will briefly discuss several material systems which make possible generation of phase states. First, let us consider an atomic system. Use of a four-state atomic system with a quantum interference, i.e., electromagnetically induced transparency, makes it possible to absorb two photons but will not absorb one photon [12]. The second candidate is a system of single exciton and biexciton, e.g., in CuCl crystal [13,14]. This system has an advantage over atomic gases as the density of oscillators is much higher than the gas. As long as the two-photon Rabi frequency

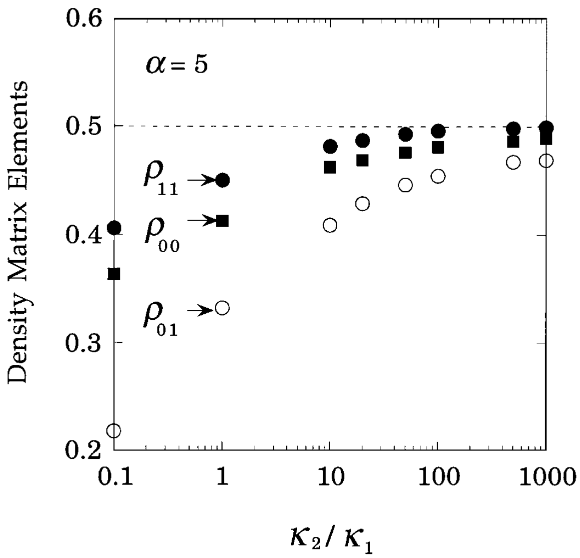

FIG. 4. Density matrix elements at the optimum interaction time (realizing a maximum value of ρ_{01}) as a function of κ_2/κ_1.

is smaller than the relaxation rate of the biexciton, the adiabatic elimination of electronic states is justified, and the present theory is applicable to the biexciton system. We can show for both systems that TPA coefficient $\alpha^{(2)}$ is chosen to be larger than the linear one $\alpha^{(1)}$ even when the light pulse contains two or three photons, as long as the beam area is less than 10^{-6} cm^2 and the laser pulse width less than 10^{-11} s. Third, some bulk semiconductors also look promising to get qubit pulse trains. The linear absorption coefficient $\alpha^{(1)}$ obeys Urbach rule below the exciton level and decreases very sharply from 10^5 cm^{-1} to 1 cm^{-1} within 10 meV change of incident photon energy, e.g., in CdS [15] and CdSe [16]. Even when we choose the incident laser pulse at such a frequency as $\alpha^{(1)}(\omega) < 1$ cm^{-1} just below the exciton peak, we can make use of the one-photon resonant enhancement of $\alpha^{(2)}$ due to the large exciton oscillator strength.

Finally, we will discuss a detection of the phase state and its application. The optical homodyne with a phase-locked-loop oscillator (OHPLL) is an ideal detector for the phase state [17,18]. The positive operator valued measure for the OHPLL becomes $P(\phi) = \frac{1}{2\pi}|\phi\rangle\langle\phi|$, where $|\phi\rangle = |0\rangle + e^{i\phi}|1\rangle$ [18]. If we consider the communication protocol that the state $|0\rangle + |1\rangle$ is detected if the measurement result lies in the interval $0 \pm \theta$ and that the state $|0\rangle - |1\rangle$ is detected if the measurement result lies in the interval $\pi \pm \theta$, then the error rate P_e is estimated by $P_e = 2\frac{1}{2\pi}\int_{\pi-\theta}^{\pi}(1+\cos\phi)\,d\phi \sim \frac{\theta^3}{6\pi}$, $(\theta \ll 1)$ and the transmission rate T is given by $T = 2\frac{1}{2\pi}\int_0^{\theta}(1+\cos\phi)\,d\phi \sim \frac{2\theta}{\pi}$. For example, we have $P_e = 5\times10^{-5}$ and $T = 6.4\%$ for $\theta = 0.1$, and $P_e = 4\times10^{-4}$ and $T = 13\%$ for $\theta = 0.2$.

A quantum cryptography using this phase state has advantages of high bit rate and security over a prototype system using an attenuated laser beam. In our scheme, Bob can select one of the two bases, $(|0\rangle \pm |1\rangle)/\sqrt{2}$ or $(|0\rangle \pm i|1\rangle)/\sqrt{2}$. This can be done by the phase modulation of the initial coherent state which is input to the two-photon absorber. Alice can independently set up her OHPLL receiver to measure either one of these two bases. She can detect the signal phase (0 or π) with a high efficiency only if her receiver setup matches Bob's base. In this way, the standard protocol is applied to our case.

In conclusion, we have proposed a new method to produce a regular series of qubit states with arbitrary phase by using two-photon absorption. This series of qubits has several advantages so that this will be useful for future quantum communication systems.

[1] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University, Cambridge, England, 1995), and references therein.
[2] A. Imamoglu and Y. Yamamoto, Phys. Rev. Lett. **72**, 210 (1994).
[3] J. Kim, O. Benson, H. Kan, and Y. Yamamoto, Nature (London) **397**, 500 (1999).
[4] A. Imamoglu, H. Schmidt, G. Woods, and M. Deutsh, Phys. Rev. Lett. **79**, 1467 (1997).
[5] D. T. Pegg, S. M. Barnett, and L. S. Phillips, J. Mod. Opt. **44**, 2135 (1997).
[6] L. Davidovich, A. Maali, M. Brune, J. M. Raimond, and S. Haroche, Phys. Rev. Lett. **71**, 2360 (1993).
[7] K. M. Cheri, C. Saavedra, P. Törmä, J. I. Cirao, and P. Zoller, Phys. Rev. A **58**, R2627 (1998); Q. A. Turchette, C. J. Hood, W. Lange, H. Mabuchi, and H. J. Kimble, Phys. Rev. Lett. **75**, 4710 (1995).
[8] M. S. Zubairy and J. J. Yeh, Phys. Rev. A **21**, 1624 (1980).
[9] H. Ezaki, G. S. Agarwal, and E. Hanamura, Opt. Commun. **138**, 65 (1997).
[10] R. R. Puri and R. K. Bullough, J. Opt. Soc. Am. B **5**, 2021 (1988).
[11] H. J. Charmichael, *An Open Systems Approach to Quantum Optics* (Springer-Verlag, Berlin, 1993); J. Dalibard, Y. Castin, and K. Molmer, Phys. Rev. Lett. **68**, 580 (1992).
[12] S. E. Harris and Y. Yamamoto, Phys. Rev. Lett. **81**, 3611 (1998).
[13] E. Hanamura, Solid State Commun. **12**, 951 (1973).
[14] G. M. Gale and A. Mysyrowicz, Phys. Rev. Lett. **32**, 727 (1974).
[15] D. L. Dexter, in *Proceedings of the 1958 Interernational Conference on Semiconductors, Rochester* (Pergamon, New York, 1959), p. 473.
[16] R. Ludeke and W. Paul, in *Proceedings of the 7th International Conference on II-VI Semiconducting Compounds, Province* (Benjamin, New York, 1967), p. 123.
[17] Y. Yamamoto *et al.*, *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1990), Vol. 28, p. 150.
[18] H. M. Wiseman, Phys. Rev. Lett. **75**, 4587 (1995).