Design of Many-Atom Cavity QED Systems for Strong Two-Photon Nonlinearity

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We propose a new scheme for strong two-photon nonlinearity (TPN) by cavity QED systems with many three-level atoms, although the TPN due to the saturation effect of two-level atoms generally declines with an increase in the number of atoms. The reduction in anharmonicity by the increase in the number of atoms can be overcome if the system satisfies particular criteria. The new scheme is significant for realizing TPN devices using solid-state materials because it is generally difficult to control the number of atoms, which are few, introduced in the device.

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The study of nonlinear effects of photon number states is crucial to understanding the fundamental quantum properties of photons and to realizing quantum information devices, which use photons as information carriers. The optical nonlinearity via a medium such as an atom, a molecule, and a solid-state material is essentially small if the photon number is of the order of unity. However, the nonlinearity can be enhanced by placing the nonlinear medium in a cavity because the electric field inside a cavity and the coupling rate between a medium and a photon are amplified; in fact, the nonlinearity of a two-level atom in a cavity has been experimentally measured as a nonlinear phase shift [1]. Nowadays, the enhanced two-photon nonlinearity (TPN) of cavity QED systems is actively studied from both a fundamental viewpoint [2–5] and an application viewpoint for quantum information technology, such as a source of entangled photon pairs [6,7] and a quantum phase gate [8].

The two-photon nonlinearity in an atom-cavity system attains a maximum for N (number of atoms) $= 1$ if the nonlinear medium consists of independent two-level atoms; however, the TPN easily declines with an increase in the number of atoms. This is because a single atom is most strongly saturated by a single photon, while the atom system cannot be saturated by only one photon if N is large [4]. This nature is unfavorable for the implementation of devices because it is not easy to control the number of introduced atoms (molecules, quantum dots) in a regime with few numbers of atoms when devices based on solid-state technology are produced. However, if we consider three-level atoms as nonlinear media, this situation can be drastically changed. In this Letter, we propose a new process for TPN, where a strong nonlinearity is observed even for large N if the atom-cavity system satisfies particular criteria. This finding is a vital step toward conducting practical experiments on TPN and the development of quantum information technology.

We consider N three-level atoms in a one-sided cavity, as shown in Fig. 1. Each atom has three states—the ground state $|g\rangle$, the first excited state $|a_i\rangle$, and the second excited state $|b_i\rangle$. The energies of $|g\rangle$, $|a_i\rangle$, and $|b_i\rangle$ are $\omega_g = 0$, ω_a, and $\omega_b = 2\omega_a - \Delta_{ab}$, respectively. The transitions between $|g\rangle$ and $|a\rangle$ and between $|a\rangle$ and $|b\rangle$ are coupled to a cavity photon by the coupling rates g_a and g_b, respectively. ω_c denotes the energy of a cavity photon, and the resonant condition $\omega_c = \omega_a$ is assumed in this study. The photons present inside and outside the cavity are linked by a partially transmitting mirror with a photon loss rate κ. The entire system comprising the N three-level atoms and internal and external photon fields of the one-sided cavity can be regarded as a one-dimensional system [2,4]. We can assume that a pair of input photons comprising two Gaussian pulses is introduced into the cavity through the right-hand mirror from the negative direction and exits through the same mirror along the positive direction via the interaction with the N-atom-cavity QED system placed at the origin $\mathbf{r} = 0$. The Hamiltonian is given as $H = \sum_{i=1}^{N}(\omega_a|a_i\rangle\langle a_i| + \omega_b|b_i\rangle\langle b_i|) + \omega_c c^\dagger c + \sum_{i=1}^{N} g_a(c^\dagger|g\rangle\langle a_i| + |a_i\rangle\langle g|c) + \sum_{i=1}^{N} g_b(c^\dagger|a_i\rangle\langle b_i| + |b_i\rangle\times\langle a_i|c) + \sum_{\mathbf{k}}|\mathbf{k}|d_{\mathbf{k}}^\dagger d_{\mathbf{k}} + \sum_{\mathbf{k}}(\kappa/L)^{1/2}(c^\dagger d_{\mathbf{k}} + d_{\mathbf{k}}^\dagger c)$. We set $\hbar$ and the light velocity to unity. $c^\dagger$ (c) and $d_{\mathbf{k}}^\dagger$ ($d_{\mathbf{k}}$) are the creation (annihilation) operators of a cavity photon and an external photon with a one-dimensional wave number $\mathbf{k}$,

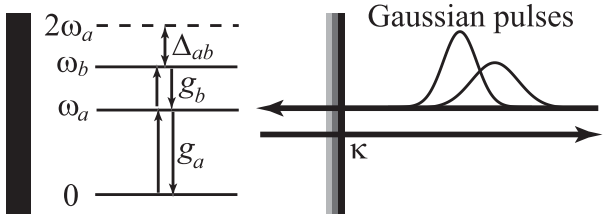

FIG. 1. Schematic view of a three-level atom in a cavity. The rate of photon loss of a one-sided cavity mirror is κ. The two pulses of a pair of input photons are Gaussian. $|g\rangle$, $|a\rangle$, and $|b\rangle$ are the states of the three-level atom. ω_a and $\omega_b = 2\omega_a - \Delta_{ab}$ are the energies of $|a\rangle$ and $|b\rangle$, respectively. g_a and g_b are the coupling rates of the $|g\rangle$ to $|a\rangle$ and $|a\rangle$ to $|b\rangle$ transitions with a photon, respectively.

respectively. L is the size of the complete system. Practical systems possess other characteristics such as damping of atoms, inhomogeneity of atomic energy levels, and so on. These characteristics may affect the TPN; however, we ignore them in order to clarify the upper limit of an ideal TPN.

We consider optical processes and a measure of the TPN along the lines of Ref. [4]. The initial state vectors of a single input photon and a pair of input photons are given by $|\Phi_{\rm in}^{\mu}\rangle = \int_L d\mathbf{r}\phi_\mu(\mathbf{r}-\mathbf{r}_0)d_{\mathbf{r}}^\dagger|0\rangle$ and $|\Psi_{\rm in}^{(2)}\rangle = 2^{-1/2}\int_L\int_L d\mathbf{r}_1 d\mathbf{r}_2\psi(\mathbf{r}_1-\mathbf{r}_0,\mathbf{r}_2-\mathbf{r}_0)d_{\mathbf{r}_1}^\dagger d_{\mathbf{r}_2}^\dagger|0\rangle$, respectively. The wave functions of photons are given by $\phi_\mu(\mathbf{r}) = [2/(\pi d_\mu)]^{1/4}\exp(-\mathbf{r}^2/d_\mu^2 + i\mathbf{k}_\mu\cdot\mathbf{r})$ and $\psi(\mathbf{r}_1,\mathbf{r}_2) = N^{(2)}[\phi_a(\mathbf{r}_1-\Delta_r/2)\phi_b(\mathbf{r}_2+\Delta_r/2) + \phi_a(\mathbf{r}_2-\Delta_r/2)\phi_b(\mathbf{r}_1+\Delta_r/2)]$, where $N^{(2)}$ is the normalization factor. $d_{\mathbf{r}}^\dagger = L^{-1/2}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{r}}d_{\mathbf{k}}^\dagger$ is a creation operator of an external photon at a position $\mathbf{r}$. k_μ and d_μ are the energy and pulse duration of an input photon μ ($\mu = a$ or b). The positions of the input photons at $t=0$ are $\mathbf{r}_0 = -L/2$. The two input photons a and b have different energies and pulse durations. The two input photons are separated from each other by distance Δ_r. The output photon states attained by the interaction with the N-atom-cavity QED system can be calculated by the Schrödinger equation $|\psi_{\rm out}(t)\rangle = \exp(-iHt)|\psi_{\rm in}(t=0)\rangle$. The state vector of a single output photon for a single input photon $|\Phi_{\rm in}^{\mu}\rangle$ and the state vector of a pair of output photons for a pair of input photons $|\Psi_{\rm in}^{(2)}\rangle$ are represented as $|\Phi_{\rm out}^{\mu}(t)\rangle = \int_L d\mathbf{r}\bar{\phi}_\mu(\mathbf{r})d_{\mathbf{r}}^\dagger|0\rangle$ and $|\Psi_{\rm NL}^{(2)}(t)\rangle = 2^{-1/2}\int_L\int_L d\mathbf{r}_1 d\mathbf{r}_2\times\bar{\psi}_{\rm nl}(\mathbf{r}_1,\mathbf{r}_2)d_{\mathbf{r}_1}^\dagger d_{\mathbf{r}_2}^\dagger|0\rangle$, respectively. Furthermore, we define another process referred to as the linear two-photon process. This can be expressed by only single-photon processes. A linear output state for a pair of input photons $|\Psi_{\rm in}^{(2)}\rangle$ is expressed as $|\Psi_L^{(2)}(t)\rangle = 2^{-1/2}\int_L\int_L d\mathbf{r}_1 d\mathbf{r}_2\times\bar{\psi}_{\rm lin}(\mathbf{r}_1,\mathbf{r}_2)d_{\mathbf{r}_1}^\dagger d_{\mathbf{r}_2}^\dagger|0\rangle$ where $\bar{\psi}_{\rm lin}(\mathbf{r}_1,\mathbf{r}_2) = N^{(2)}[\bar{\phi}_a(\mathbf{r}_1)\bar{\phi}_b(\mathbf{r}_2) + \bar{\phi}_a(\mathbf{r}_2)\bar{\phi}_b(\mathbf{r}_1)]$. We define a measure of the TPN as $\alpha = \langle\Psi_L^{(2)}(t_f)|\Psi_{\rm NL}^{(2)}(t_f)\rangle$, where $t_f = L$. $|\alpha|$ and $|\arg(\alpha)|$ indicate the fidelity of a two-photon wave function and the nonlinear phase shift of the two-photon state via a nonlinear process, respectively. The optimal condition for a quantum logic gate is $|\alpha| = 1$ and $|\arg(\alpha)| = \pi$.

We consider the criteria for designing the N-atom-cavity QED system in order to obtain a strong TPN. The high-Q-cavity regime is assumed in this Letter. In this case, new eigenstates are created by the excited states $\{|a_i\rangle, |b_i\rangle\}$ of N atoms and the cavity-photon state $|c\rangle$. The peculiar properties of this energy structure provide the conditions for a strong TPN, even for the case of large N. In the case of one-photon incidence, the atom-cavity system is constructed with two bases, namely, $|A\rangle = N^{-1/2}\sum_{i=1}^N|a_i\rangle$ and $|c\rangle$, and one-particle excited states ($1P$ states) are described as superpositions $|1P_\pm\rangle = 2^{-1/2}(|A\rangle \pm |c\rangle)$. The eigenenergies and widths of $|1P_\pm\rangle$ are $\omega_{1P_\pm} \approx \omega_a \pm N^{1/2}g_a$ and $\Delta\omega_{1P_\pm} \approx \kappa/4$, respectively. In the case of two-photon incidence, two-particle excited states ($2P$ states) are superposition states consisting of four bases, namely, $|B\rangle = N^{-1/2}\sum_{i=1}^N|b_i\rangle$, $|AA\rangle = \{2/[N(N-1)]\}^{1/2}\sum_{i,j(i<j)}|a_i\rangle|a_j\rangle$, $|A\rangle|c\rangle$, and $|2c\rangle$, whose coefficients change with various parameters of the N-atom-cavity QED system. Figure 2 shows the energy levels of the $2P$ states $|2P_1\rangle$, $|2P_2\rangle$, $|2P_3\rangle$, and $|2P_4\rangle$ (solid lines). The horizontal broken line denotes the energy level of $|B\rangle$. The diagonal broken lines denote the energy levels of the superposition states $|2H_1\rangle$ and $|2H_2\rangle$ consisting of $|AA\rangle$, $|A\rangle|c\rangle$, and $|2c\rangle$. Their energy levels change according to the system parameter $[2(2N-1)]^{1/2}g_a/\Delta_{ab}$. The optimal $2P$ state for a strong TPN has sufficient anharmonicity and strong coupling to the photons. The origins of the coupling to photons are $|2H_1\rangle$ and $|2H_2\rangle$; therefore, the optimal $2P$ state must have $|2H_1\rangle$ or $|2H_2\rangle$ as a component. However, $|2H_1\rangle$ and $|2H_2\rangle$ are harmonic and cannot cause nonlinearity. The origin of the anharmonicity is $|B\rangle$; therefore, the optimal $2P$ state must have also $|B\rangle$ as a component. In short, the optimal $2P$ state for obtaining a strong TPN must be a superposition $w_1|B\rangle + w_2|2H_1\rangle$ with optimal weights w_1 and w_2. The $2P$ states with both $|B\rangle$ and $|2H_1\rangle$ as components are $|2P_1\rangle$ and $|2P_2\rangle$. We find two conditions for realizing the optimal $|2P_1\rangle$ and $|2P_2\rangle$ states, as follows:

$$N \approx 2^{-1}[1 + 2^{-1}(\Delta_{ab}/g_a)^2] \tag{1}$$

and

$$\kappa < g_b. \tag{2}$$

Equation (1) is the condition for the optimal weights w_1 and w_2. This situation corresponds to the area where the lines of $|B\rangle$ and $|2H_1\rangle$ cross each other in Fig. 2. In this area, these two states interact with each other and split into $|2P_1\rangle$ and $|2P_2\rangle$ levels, shifting from the harmonic state

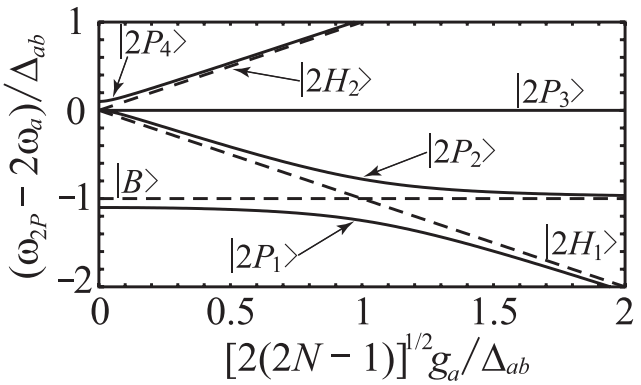

FIG. 2. Dependence of $2P$-state energy levels on the system parameter $[2(2N-1)]^{1/2}g_a/\Delta_{ab}$. $|2P_1\rangle$, $|2P_2\rangle$, $|2P_3\rangle$, and $|2P_4\rangle$ are the $2P$ states. $|B\rangle$ is the anharmonic second excited state of N atoms, and $|2H_1\rangle$ is a harmonic state that is a superposition of $|AA\rangle$, $|A\rangle|c\rangle$, and $|2c\rangle$.

$|2H_1\rangle$. Equation (2) is the condition for $|2P_1\rangle$ and $|2P_2\rangle$ to obtain sufficient anharmonicity. The width of the level splitting is of the order of g_b, while the level width of each energy level of $|2P_1\rangle$ and $|2P_2\rangle$ is of the order of the photon loss rate κ. Thus, Eqs. (1) and (2) specify the criteria for designing a cavity QED system for obtaining a strong TPN. The meanings of these criteria are as follows. (i) If the rate of first transition between the ground state and the first excited state of an atom (g_a) is considerably smaller than Δ_{ab}, a large number of atoms ($N \gg 1$) provides the maximum TPN. (ii) If the cavity quality factor is larger than the inverse of the coupling rate (g_b) between the first and the second excited states of an atom, a strong TPN can be obtained. The essence of this new scheme is that the anharmonicity of a system can be enhanced by using cooperative effects of N atoms via the photon field inside a cavity. The states $|A\rangle$, $|B\rangle$, and $|AA\rangle$ are the collective states which are generally known as the Dicke-superradiant states, furthermore, they are coupled to a cavity-photon state $|c\rangle$. Therefore, in the appropriate conditions, the anharmonicity of the entire system can be enhanced by controlling the energy-level structure of an atom-cavity system. Before we discuss the materials that can be used to realize these conditions, we provide some numerical demonstrations.

Here, we calculate the TPN phase shift. We set the parameters of atoms and the cavity arbitrarily as $\Delta_{ab} = 3$, $g_b = 2$, and $\kappa = 0.2$; however, the values of g_b and κ satisfy Eq. (2). We use an arbitrary unit for energy. The role of the photon a is to excite the $|1P_-\rangle$ state so that the energy k_a of the photon a is resonant to $|1P_-\rangle$, and the pulse duration d_a is decided by the level width of $|1P_-\rangle$ as $k_a = \omega_{1P-} = \omega_a - N^{1/2}g_a$ and $d_a = 3/\kappa$. Moreover, the photon a is incident before the photon b with respect to the excitation to the $2P$ state, and we set $\Delta_{\mathbf{r}} = 0.6d_b$. In order to obtain the most optimal TPN, we must use the two-photon process by using the $|2P_1\rangle$ and $|2P_2\rangle$ states. For this purpose, the sum of two-photon energies $k_a + k_b$ should be near the area of the level splitting of $|2P_1\rangle$ and $|2P_2\rangle$ shown in Fig. 2, and the pulse duration d_b of the photon b should be decided by the level widths of the $2P$ states and the width of the level splitting. In this Letter, we set $k_a + k_b = \omega_b + 0.58g_b$ and $d_b = 3.3/\kappa$. These values of parameters of the two input photons are most optimal for realizing the maximum TPN phase shift. Figure 3 shows the dependence of $|\arg(\alpha)|$ on N. Three cases of doped atoms are considered: namely, (i) $g_a = 1$, (ii) $g_a = 0.1$, and (iii) $g_a = 0.01$. Interestingly, we find that if $g_a \ll \Delta_{ab}$, the optimal TPN phase shift occurs for large N, which is determined by Eq. (1). Further, as observed in the inset of Fig. 3, if g_a is sufficiently small, the large shift is maintained for a wide range of N. This implies that we are required to control only the order of N in order to obtain a large TPN phase shift; however, this is in contrast to the fact that an accurate control of N is required for the large TPN phase shift when

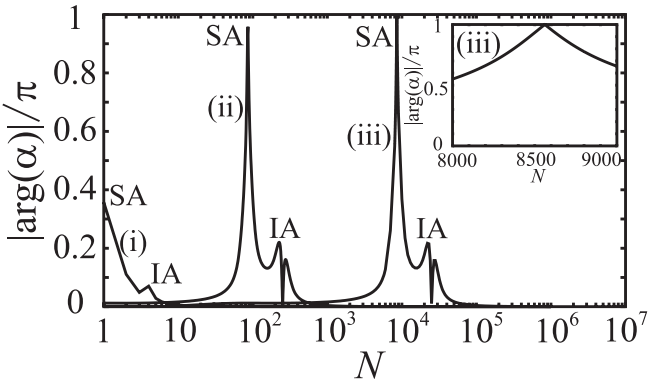

FIG. 3. Dependence of the two-photon nonlinear phase shift $|\arg(\alpha)|$ on the number of atoms (N) doped in a cavity. Three cases, namely, (i) $g_a = 1$, (ii) $g_a = 0.1$, and (iii) $g_a = 0.01$, are shown. We found two kinds of peaks, SA and IA. The other parameters are $\Delta_{ab} = 3$, $g_b = 2$, and $\kappa = 0.2$.

g_a is of the order of unity. From cases (ii) and (iii) shown in Fig. 3, we find that two peaks—the saturation of absorption (SA) and the induced absorption (IA)—and two optimal numbers of atoms exist. The dynamics of a system are investigated in order to clarify the details of the SA and IA processes. The dynamics of a system in case (iii) shown in Fig. 3, $g_a = 0.01$, are shown in Fig. 4. Figures 4(a) and 4(b) are the case of (SA) $N = 8600$ and (IA) $N = 25\,100$. The curves (I), (II), and (III) show the occupation rates of the external two-photon state, $|1P_-\rangle$, and $|2P_2\rangle$, respectively. Firsty, we discuss the SA peak shown in Fig. 4(a). $|1P_-\rangle$ is excited; however, $|2P_2\rangle$ is not excited. In this case, the energies of the two input photons are equal to $k_a = k_b = \omega_a - N^{1/2}g_a$. We conclude that the increase in the TPN phase shift at the SA peak is caused by the saturation of absorption. The saturation occurs because the level with the energy of $k_a + k_b$ is not formed by the level splitting. Second, we discuss the IA peak shown in Fig. 4(b). Both $|1P_-\rangle$ and $|2P_2\rangle$ are excited and relaxed in succession; this demonstrates the transition between $|1P_-\rangle$ and $|2P_2\rangle$. We

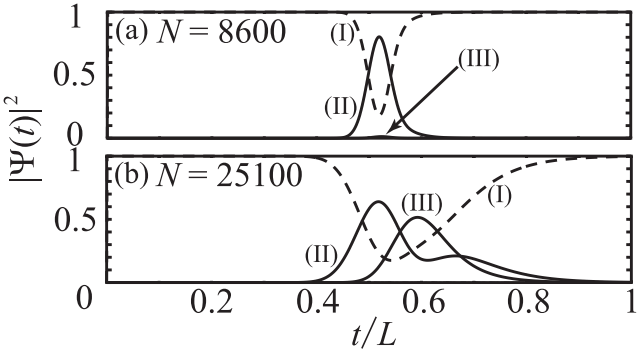

FIG. 4. Time evolution of occupation rates of (I) the external two-photon state, (II) $|1P_-\rangle$, and (III) $|2P_2\rangle$. Case (a) $N = 8600$ corresponds to the SA peak and case (b) $N = 25100$ corresponds to the IA peak of (iii) $g_a = 0.01$.

conclude that the increase in the TPN phase shift at the IA peak is caused by the induced absorption from $|1P_-\rangle$ to $|2P_2\rangle$. Thus, we have clarified the existence of two optimal numbers of doped atoms, which correspond to individual nonlinear processes—the saturation of absorption and the induced absorption—in order to realize a strong TPN. We have also confirmed the trade-off relation between the fidelity $|\alpha|$ and the phase shift $|\arg(\alpha)|$, which is similar to that discussed in Ref. [4].

Finally, we discuss the material that satisfies the criteria and realizes a strong TPN even if N is large. The favorable conditions are summarized as follows. First, the material must be a three-level system with the anharmonicity Δ_{ab}. Second, the transition between the ground state $|g\rangle$ and the first excited state $|a\rangle$ is almost forbidden. Third, the photon loss rate κ of the cavity is smaller than the coupling rate g_b of the transition between the first excited state $|a\rangle$ and the second excited state $|b\rangle$. The following are the examples of such materials satisfying these conditions. Molecules such as pyrene have a three-level structure [9]. The S_0-S_1 ($|g\rangle$-$|a\rangle$) transition and the S_1-S_n ($|a\rangle$-$|b\rangle$) transition are observed at two different energies separated by $\Delta_{ab} = 50$ meV [10]. The S_0-S_1 transition is almost forbidden when $g_a \approx 10^{-2}$ meV, and N_0 becomes approximately 10^6 (here, N_0 denotes the number of atoms for the optimal TPN). However, the S_1-S_n transition is allowed, and Eq. (2) is satisfied if $\kappa < 10^{-1}$ meV. A quantum dot that confines the exciton $|a\rangle$ and the biexciton $|b\rangle$ is another candidate. The coupling rates are calculated as $g_a \propto \int dr \psi_{\mathrm{ex}}(r) u_c(r) \psi_g(r)$ and $g_b \propto \int dr \psi_{\mathrm{bx}}(r) u_c(r) \psi_{\mathrm{ex}}(r)$, where $\psi_g(r)$, $\psi_{\mathrm{ex}}(r)$, and $\psi_{\mathrm{bx}}(r)$ are ground-state, excitonic, and biexcitonic wave functions, respectively, of center-of-mass motion, and $u_c(r)$ is the cavity mode function. If we consider a λ cavity, where $u_c(r)$ is an odd function with a node at the center of the cavity and $\psi_{\mathrm{ex}}(r)$ is an even function, we can arbitrarily control the relation between g_a and Δ_{ab} in order to realize the large N_0 by a slight shift in the position of the quantum dot from the center. Further, Eq. (2) is satisfied if $\kappa < 10^{-2}$ meV in the case of a CuCl quantum dot with its size of the order of nanometers. In both the cases, the effect of inhomogeneity should be considered. Although this effect is not included in the present model, we can expect that a strong coupling between a quantum dot (molecule) and a photon can overcome the problem due to the inhomogeneity of energies; the coupling rate of $1P$ states is enhanced by $N^{1/2}$. If N is sufficiently large, the level splitting of $1P$ states is expected to be so large that the increase in level width due to the inhomogeneity does not affect the present mechanism significantly. If the inhomogeneity of the biexcitonic level is large, it might degrade the anharmonicity of the system. However, the directions of the shifts of the excitonic and biexciton binding energies due to the size change of the quantum dot are opposite to each other; hence, the oppositely directed shifts cancel each other to some extent and the inhomogeneity of the biexcitonic level is reduced. A detailed analysis of this problem will be discussed in the next publication. The problem of inhomogeneity can be avoided if the solid-state material is doped with rare-earth ions because they have extremely good homogeneity. For example, certain levels of Er^{3+} and Tm^{3+} provide an ideal three-level system [11], which results in approximately $N_0 \approx 10^{11}$, and Eq. (2) is satisfied if $\kappa < 10^{-4}$ meV. If we dope the ions in a high-Q spherical cavity, this condition can be realized [12]. By employing these materials, a cavity QED system satisfying Eqs. (1) and (2) can be realized. The criteria proposed in this study can be applied widely and can aid the realization of a two-photon quantum device using solid-state materials. We expect that this study will stimulate the study of TPN by practical experiments using solid-state materials.

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