Three-qubit phase gate based on cavity quantum electrodynamics

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We describe a three-qubit quantum phase gate which is implemented by passing a four-level atom in a cascade configuration initially in its ground state through a three-mode optical cavity. The three qubits are represented by the photons in the three modes of the cavity. Under appropriate detunings, coupling coefficients, and interaction times, this system exhibits like a three-qubit quantum phase gate. Such a quantum phase gate has potential applications in quantum search.

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I. INTRODUCTION

Quantum computing provides a novel way to carry out certain computations much faster than the conventional Turing machines. This is, in general, accomplished by performing specific unitary transformations on a set of quantum bits (qubits) followed by measurement. Quantum computing has been shown to provide an efficient means for computation in two distinct problems. Shor's algorithm to factorize a number can provide a polynomial time cost instead of subexponential time cost by a classical computer. Since many cryptographic systems are based on the complexity of integer factorizing, Shor's algorithm triggered a tremendous enthusiasm for building a quantum computer. Another example of the ability of quantum computation is the quantum search of an object in an unsorted database containing N elements. A quantum computing algorithm allows a search to be carried out in, on average, $O(\sqrt{N})$ steps, while it takes $O(N)$ steps to do so in classical computation. Many physical systems have been proposed as candidates for quantum computer implementation, such as linear ion traps [1], liquid-state nuclear magnetic resonance (NMR) [2], and cavity QED systems [3,4].

There are three requirements for implementing a quantum computer: Efficient manipulation and read out of an individual qubit's state, strong coupling between qubits (fast gate operation) and weak coupling to environment (slow decoherence), and scalability. The scheme with qubits based on photon number states in a strong coupling optical cavity offers itself as a good choice [5,6].

The basic element of a computer is the logic gate, either in a classical computer or a quantum computer. The key point of quantum computer architecture is therefore to find an effective physical realization of quantum logic gates. Moreover, from quantum network theory, any entangling two-qubit gates assisted by a one-qubit gate is universal for quantum computation [7]. The two-qubit quantum phase gate is one of them. Particularly, a quantum phase gate can be directly used in the implementation of Grover's search algorithm [8], quantum Fourier transformation [9], quantum error correction [10], and arbitrary superposed state preparation [11]. In the earlier work of Zubairy *et al.* [6], a scheme to implement a two-qubit quantum phase gate and one-qubit unitary operation implementation based on cavity QED was described. They choose the Fock states $|0\rangle$ and $|1\rangle$ of a high Q cavity mode as the two logical states of a qubit. The phase gate is accomplished by passing a ground state three-level atom through the cavity and the one-qubit unitary operation is realized by passing a two-level atom through the cavity accompanied by shining in a classical light pulse. In this paper, we extend their method to the implementation of a three-qubit gate.

It has been shown that a three-qubit quantum phase gate, and more generally a multiqubit quantum phase gate, can lead to faster computing. For example, it plays a key role in the realization of quantum error correction [10] and the implementation of Grover's search algorithm for eight objects [12]. However, according to Diao *et al.* [13], a three-qubit quantum phase gate needs a network of four one-bit quantum gates and five two-qubit quantum phase gates. The realization of a three-qubit quantum phase gate can be considerably simplified.

In many schemes to implement quantum logic gates, the qubits are represented by different systems, such as the internal state of an atom and the quantum states of the optical field. However, the experimental realization of a typical quantum algorithm may require each qubit to be treated equally. In our scheme based on cavity QED, the three qubits are represented by three different modes of the field inside the cavity. An earlier NMR based three-qubit quantum phase gate implementation was reported by Zhang *et al.* [14].

II. THREE-QUBIT QUANTUM PHASE GATE

A two-qubit quantum phase gate with phase η can be described by the operator

$$Q_\pi = |0_1,0_2\rangle\langle 0_1,0_2| + |1_1,0_2\rangle\langle 1_1,0_2| + |0_1,1_2\rangle\langle 0_1,1_2| + e^{i\phi}|1_1,1_2\rangle\langle 1_1,1_2|. \tag{1}$$

Similarly, the transformation for a three-qubit quantum phase gate can be defined via

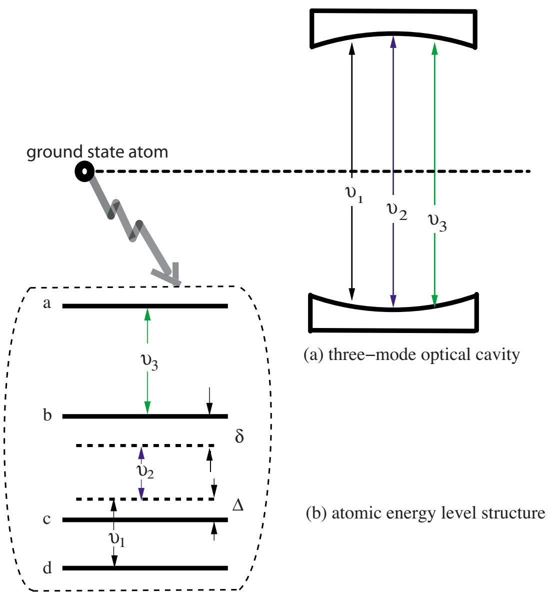

FIG. 1. (Color online) Scheme of three-qubit phase gate, where (a) ν_1, ν_2, and ν_3 are modes of the three-mode high Q cavity. $\omega_{cd}-(\nu_1/2\pi)=\Delta$, $\omega_{cd}+\omega_{bc}-(\nu_1/2\pi)-(\nu_2/2\pi)=\delta$, and $\omega_{ab}=\nu_3/2\pi$. (b) a, b, c, and d are four energy levels of the atom.

$$Q_\eta|\alpha_1,\beta_2,\gamma_3\rangle = \exp(\imath\eta\delta_{\alpha_1,1}\delta_{\beta_2,1}\delta_{\gamma_3,1})|\alpha_1,\beta_2,\gamma_3\rangle,$$

where $|\alpha_1\rangle$, $|\beta_2\rangle$, and $|\gamma_3\rangle$ represent the basis states $|0\rangle$ or $|1\rangle$ of the qubits 1, 2, and 3, respectively. Thus the quantum phase gate introduces a phase η only when all the qubits in the input state are 1. The operator Q_η can be written as

$$\begin{aligned}Q_\eta = {} & |0_1,0_2,0_3\rangle\langle 0_1,0_2,0_3| + |1_1,0_2,0_3\rangle\langle 1_1,0_2,0_3| \\ & + |0_1,1_2,0_3\rangle\langle 0_1,1_2,0_3| + |0_1,0_2,1_3\rangle\langle 0_1,0_2,1_3| \\ & + |0_1,1_2,1_3\rangle\langle 0_1,1_2,1_3| + |1_1,0_2,1_3\rangle\langle 1_1,0_2,1_3| \\ & + |1_1,1_2,0_3\rangle\langle 1_1,1_2,0_3| + \exp(\imath\eta)|1_1,1_2,1_3\rangle\langle 1_1,1_2,1_3|. \end{aligned} \quad (2)$$

In the following we discuss a cavity QED implementation of the case with $\eta=\pi$.

We consider a system as shown in Fig. 1. The atom has cascade four energy levels which are denoted by a, b, c, and d. The high Q cavity has three modes of frequencies ν_1, ν_2, and ν_3. The cavity field frequencies ν_1 and ν_2 are assumed to be far detuned from the energy differences of the first three atomic energy levels, but the sum of ν_1 and ν_2 is appropriately detuned from the energy difference of the first and the third atomic energy levels. Field mode ν_3 is chosen to be resonant with the third and fourth atomic energy levels. The detunings Δ and δ are indicated in Fig. 1. The relation between the atomic transitions and cavity modes can be described as follows:

$$\Delta = \omega_{cd} - \frac{\nu_1}{2\pi},$$

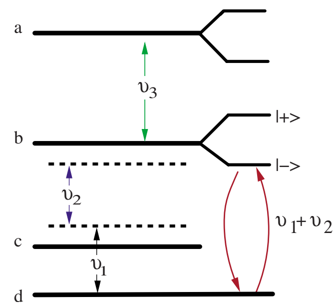

FIG. 2. (Color online) Dressed state picture based on $\nu_1+\nu_2$ two-photon transition process and the presence of ν_3 photon field.

$$\delta = \omega_{cd} + \omega_{bc} - \frac{\nu_1}{2\pi} - \frac{\nu_2}{2\pi},$$

$$\nu_3 = \omega_{ab}. \quad (3)$$

The photon number states $|0\rangle$ and $|1\rangle$ represent logic 0 and 1. All the possible cavity states are therefore given by $|0_1,0_2,0_3\rangle$, $|1_1,0_2,0_3\rangle$, $|0_1,1_2,0_3\rangle$, $|0_1,0_2,1_3\rangle$, $|1_1,1_2,0_3\rangle$, $|1_1,0_2,1_3\rangle$, $|0_1,1_2,1_3\rangle$, and $|1_1,1_2,1_3\rangle$.

We consider an atom with such an energy level structure in its ground state passing through the cavity. It is clear that $|0_1,0_2,0_3\rangle$ state is not affected by the passage of the atom. In addition, since the modes ν_1 and ν_2 are far detuned from the atomic transition frequencies ω_{cd} and ω_{bc}, respectively, the atomic state as well as the cavity field state remain unaffected when there is only one photon in any of the three cavity modes, i.e., the states $|1_1,0_2,0_3\rangle$, $|0_1,1_2,0_3\rangle$, and $|0_1,0_2,1_3\rangle$ do not change. Similarly, the cavity states $|1_1,1_2,0_3\rangle$, $|1_1,0_2,1_3\rangle$, and $|0_1,1_2,1_3\rangle$ remain unaffected. However, the situation is nontrivial when there is one photon in each of the three cavity modes. Due to the presence of a photon in mode 3, the atomic energy level b splits into two levels $|+\rangle$ and $|-\rangle$ (dynamic Stark shift), as indicated in Fig. 2. Under certain conditions, when the splitting is large enough, the two photons in mode 1 and mode 2 couple the two levels $|d\rangle$ and $|-\rangle$ via the two-photon process. The atom-field system undergoes Rabi oscillations with a frequency Ω which is determined by the coupling strength and the detuning between the atom and field modes as discussed later in this section. If the duration τ of the interaction between the field and atom satisfies the condition $\Omega\tau=(2n+1)\pi$ (n is a positive integer), we get a π phase shift on the system, i.e., $|1_1,1_2,1_3\rangle \to -|1_1,1_2,1_3\rangle$. In view of the definition in Eq. (1), this would represent a possible realization of a three-bit quantum phase gate with a π phase shift.

In the following we discuss this phase gate scheme first in a dressed state picture that brings out the physics clearly. Then we present the exact solutions and plot out different cases for comparison.

First we derive the effective Hamiltonian for our system. With the effective Hamiltonian, we are able to show how the π phase shift can be introduced for the initial state $|d\rangle \otimes |1_1,1_2,1_3\rangle$ under specific conditions.

The Hamiltonian for the system, with the dipole and rotating-wave approximations, can be written as [15,16]

$$H = H_A + H_F + H_{AF}, \tag{4}$$

where

$$H_A = \hbar(\omega_{ab}|a\rangle\langle a| + \omega_{bc}|b\rangle\langle b| + \omega_{cd}|c\rangle\langle c|), \tag{5}$$

$$H_F = \hbar\omega_1\hat{a}_1^\dagger\hat{a}_1 + \hbar\omega_2\hat{a}_2^\dagger\hat{a}_2 + \hbar\omega_3\hat{a}_3^\dagger\hat{a}_3, \tag{6}$$

$$H_{AF} = \hbar[g_1|c\rangle\langle d|\hat{a}_1 + g_2|b\rangle\langle c|\hat{a}_2 + g_3|a\rangle\langle b|\hat{a}_3 + \text{H.c.}]. \tag{7}$$

Here we have assumed that

$$g_1^{cd} = g_1^{dc} = g_1, \tag{8}$$

$$g_2^{bc} = g_2^{cb} = g_2, \tag{9}$$

$$g_3^{ab} = g_3^{ba} = g_3, \tag{10}$$

where g_k^{ij} is the Rabi frequency associated with the coupling of the field mode ν_k to the atomic transition $|i\rangle \to |j\rangle$.

In the interaction picture, the Hamiltonian can be written as

$$H_I = \hbar g_1|c\rangle\langle d|\hat{a}_1 \exp(-\imath\Delta t) + \hbar g_2|b\rangle\langle c|\hat{a}_2 \exp[-\imath(\Delta+\delta)t] + \hbar g_3|a\rangle\langle b|\hat{a}_3 + \text{H.c.} \tag{11}$$

We assume that the state for the system at time t is

$$|\Psi(t)\rangle = C_a(t)|a,n_1-1,n_2-1,n_3-1\rangle + C_b(t)|b,n_1-1,n_2-1,n_3\rangle + C_c(t)|c,n_1-1,n_2,n_3\rangle + C_d(t)|d,n_1,n_2,n_3\rangle.$$

The corresponding Schrödinger equation in the interaction picture is

$$\imath\hbar\frac{\partial|\Psi(t)\rangle}{\partial t} = H_I|\Psi(t)\rangle. \tag{12}$$

It follows that the probability amplitudes satisfy the following equations of motion:

$$\imath\hbar\dot{C}_d = \hbar g_1 C_c\sqrt{n_1}\exp(\imath\Delta t), \tag{13}$$

$$\imath\hbar\dot{C}_c = \hbar g_1 C_d\sqrt{n_1}\exp(-\imath\Delta t) + \hbar g_2 C_b\sqrt{n_2}\exp[-\imath(\Delta+\delta)t], \tag{14}$$

$$\imath\hbar\dot{C}_b = \hbar g_2 C_c\sqrt{n_2}\exp[\imath(\Delta+\delta)t] + \hbar g_3 C_a\sqrt{n_3}, \tag{15}$$

$$\imath\hbar\dot{C}_a = \hbar g_3 C_b\sqrt{n_3}. \tag{16}$$

Let $C_d = d_d$, $C_c = d_c\exp(-\imath\Delta t)$, $C_b = d_b\exp(\imath\delta t)$, and $C_a = d_a\exp\imath\delta t$. Then we can rewrite the four equations as

$$\dot{d}_d = -\imath g_1\sqrt{n_1}d_c, \tag{17}$$

$$\dot{d}_c = -\imath g_1\sqrt{n_1}d_d - \imath g_2\sqrt{n_2}d_b + \imath\Delta d_c, \tag{18}$$

$$\dot{d}_b = -\imath g_2\sqrt{n_2}d_c - \imath g_3\sqrt{n_3}d_a - \imath\delta d_b, \tag{19}$$

$$\dot{d}_a = -\imath g_3 d_b\sqrt{n_3} - \imath\delta d_a. \tag{20}$$

We assume that

$$\dot{d}_c = 0, \tag{21}$$

i.e., there is no transition to the $|c\rangle$ state. We can then solve for d_c from Eq. (18) and substitute the resulting solution into Eqs. (17), (19), and (20). The new system evolution equations are equivalent to the case that we have the following Hamiltonian in the interaction picture:

$$H_I' = H_{I1} + H_{I2} + H_{I3}, \tag{22}$$

$$H_{I1} = \frac{\hbar g_1^2}{\Delta}\hat{a}_1^\dagger\hat{a}_1|d\rangle\langle d| + \frac{\hbar g_2^2}{\Delta}\hat{a}_2^\dagger\hat{a}_2|b\rangle\langle b|, \tag{23}$$

$$H_{I2} = \frac{\hbar g_2 g_1}{\Delta}[\hat{a}_1^\dagger\hat{a}_2^\dagger|d\rangle\langle b|\exp(-\imath\delta t) + \hat{a}_1\hat{a}_2|b\rangle\langle d|\exp(+\imath\delta t)], \tag{24}$$

$$H_{I3} = \hbar g_3(\hat{a}_3^\dagger|b\rangle\langle a| + \hat{a}_3|a\rangle\langle b|). \tag{25}$$

As we discussed before, we focus only on the case when the initial state is $|d,1,1,1\rangle$. We notice that, in this case, the only allowed states for the atom-field system are $|d,1,1,1\rangle$, $|c,0,1,1\rangle$, $|b,0,0,1\rangle$, and $|a,0,0,0\rangle$. It is convenient to express the Hamiltonian in terms of the symmetric and antisymmetric states (see Fig. 2):

$$|+\rangle = \frac{1}{\sqrt{2}}(|a,0,\;0,0\rangle + |b,0,0,1\rangle), \tag{26}$$

$$|-\rangle = \frac{1}{\sqrt{2}}(|a,0,0,0\rangle - |b,0,0,1\rangle). \tag{27}$$

These states represent the dynamic Stark splitting of the levels a and b. The splitting is equal to $\hbar g_3$. We consider the situation when $\delta = g_3$.

In terms of the states $|+\rangle$ and $|-\rangle$, the effective Hamiltonian $H_{I2}+H_{I1}$ in the interaction picture of H_{I3} is given by

$$H_{I\text{eff}} = \frac{\hbar g_2 g_1}{\sqrt{(2)}\Delta}[-|-\rangle\langle d,1,1,1| - |d,1,1,1\rangle\langle -| + |+\rangle\langle d,1,1,1| \times\exp(\imath 2\delta t) + |d,1,1,1\rangle\langle +|\exp(-\imath 2\delta t)] + \hbar\frac{g_1^2}{\Delta}|d,1,1,1\rangle\langle d,1,1,1| + \hbar\frac{g_2^2}{2\Delta}(|+\rangle\langle +| + |-\rangle\langle -|) - \hbar\frac{g_2^2}{2\Delta}[|-\rangle\langle +|\exp(-\imath 2g_3 t) + |+\rangle\langle -|\exp(\imath 2g_3 t)]. \tag{28}$$

When the detuning δ is large enough, we can omit the fast oscillation terms and the effective Hamiltonian is

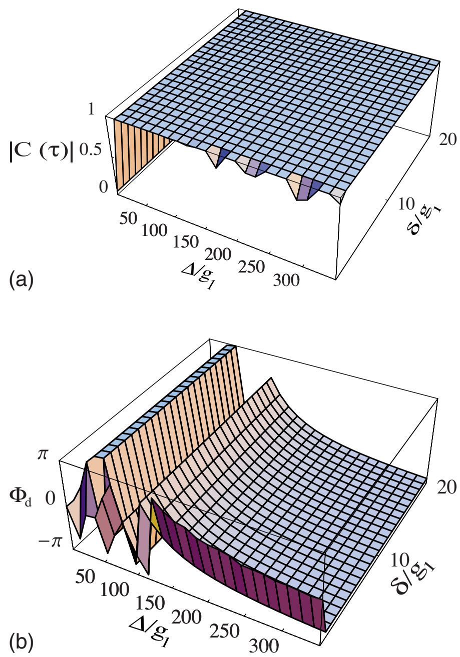

FIG. 3. (Color online) Plot of the modulus and phase of $C_d(\tau)$ $[C_d(\tau)=|C_d(\tau)|\exp(i\Phi_d)]$ as a function of Δ/g_1 and δ/g_1 under the condition that $g_1^2\tau=2n\Delta\pi$, $g_2^2\tau=2\Delta\pi$, and $n=87$. The initial state is $|d,1,1,1\rangle$.

$$H_{\mathrm{feff}}=-\frac{\hbar g_2 g_1}{\sqrt{2}\Delta}[|-\rangle\langle d,1,1,1|+|d,1,1,1\rangle\langle -|]+\hbar\frac{g_1^2}{\Delta}|d,1,1,1\rangle\langle d,1,1,1|+\hbar\frac{g_2^2}{2\Delta}(|+\rangle\langle +|+|-\rangle\langle -|). \tag{29}$$

It is easy to see that, in Eq. (29), if $g_1 \gg g_2$, the system evolution is dominated by $(\hbar g_1^2/\Delta)|d,1,1,1\rangle\langle d,1,1,1|$ term. Consequently, if $g_1^2\tau/\Delta=(2n+1)\pi$ (n is an arbitrary integer) and the initial state is $|d,1,1,1\rangle$, the final state at time τ will be $-|d,1,1,1\rangle$. This completes the description of the phase gate Q_π.

To derive the conditions more precisely, we solve the Schrödinger equation in the interaction picture using the effective Hamiltonian (29). The possible state at an arbitrary time t can be written as $C_d(t)|d,1,1,1\rangle+C_-(t)|-\rangle+C_+(t)|+\rangle$ with the initial conditions $C_d(0)=1, C_-(0)=0, C_+(0)=0$. The solutions of $C_d(t), C_-(t), C_+(t)$ are as follows:

$$C_+(t)=0, \tag{30}$$

$$C_d(t)=\frac{1}{2g_1^2+g_2^2}\left[g_2^2+2g_1^2\cos\left(\frac{-(2g_1^2+g_2^2)t}{2\Delta}\right)+2\imath g_1^2\sin\left(\frac{-(2g_1^2+g_2^2)t}{2\Delta}\right)\right], \tag{31}$$

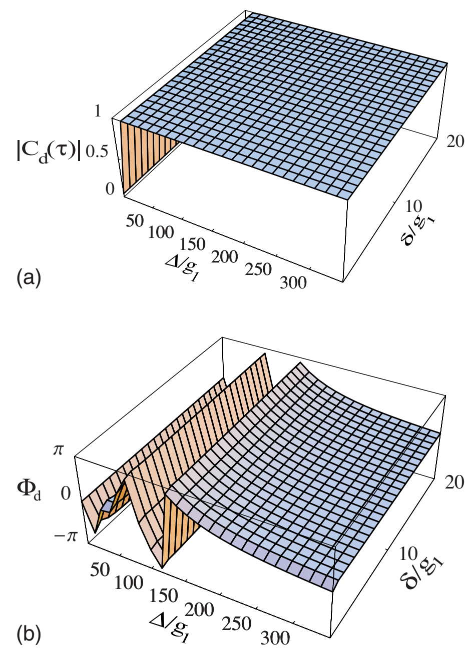

FIG. 4. (Color online) Plot of the modulus and phase of $C_d(\tau)$ $[C_d(\tau)=|C_d(\tau)|\exp(i\Phi_d)]$ as a function of Δ/g_1 and δ/g_1 under the condition that $g_1^2\tau=2n\Delta\pi$, $g_2^2\tau=2\Delta\pi$, and $n=87$. The initial state is $|d,1,1,0\rangle$.

$$C_-(t)=\frac{1}{2g_1^2+g_2^2}\left[\sqrt{2}g_1g_2-\sqrt{2}g_1g_2\cos\left(\frac{-(2g_1^2+g_2^2)t}{2\Delta}\right)-\imath\sqrt{2}g_1g_2\sin\left(\frac{-(2g_1^2+g_2^2)t}{2\Delta}\right)\right]. \tag{32}$$

In Eqs. (31) and (32), when

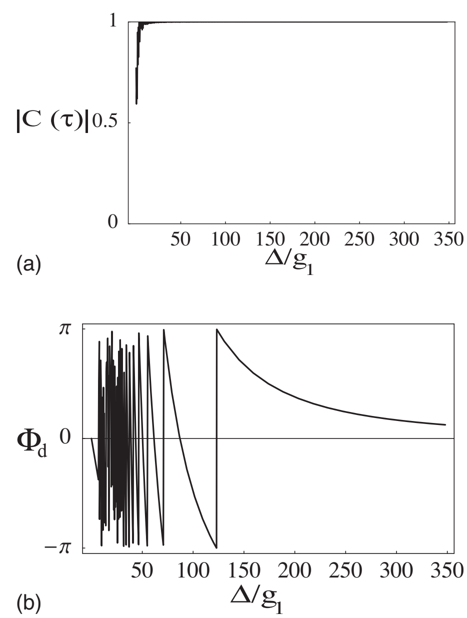

FIG. 5. Plot of the modulus and phase of $C_d(\tau)$ $[C_d(\tau)=|C_d(\tau)|\exp(i\Phi_d)]$ as the function of Δ/g_1 under the condition that $g_1^2\tau=2n\Delta\pi$, $g_2^2\tau=2\Delta\pi$, and $n=87$. The initial state is $|d,1,0,0\rangle$.

$$\frac{(2g_1^2+g_2^2)\tau}{2\Delta}=(2n+1)\pi, \quad g_1 \gg g_2, \tag{33}$$

with n being a positive integer, we have $C_d(\tau)\to -1$ and $C_-\to 0$. This implies the realization of π phase shift for the input state. In addition to the above two conditions, we also need to satisfy two more conditions, i.e.,

$$\delta = g_3, \quad \Delta \gg g_1. \tag{34}$$

In order to examine these four conditions, we plot the exact solutions for the amplitude $C_d(\tau)$ (see the Appendix) for initial states $|d,1,0,0,0\rangle$, $|d,1,1,0\rangle$, and $|d,1,1,1\rangle$ in Figs. 3–5, respectively. Here we have chosen $g_1^2\tau=2n\Delta\pi$, $g_2^2\tau=2\Delta\pi$, and $n=87$. From these figures, it is clear that when $\Delta/g_1\sim 200$ and $\delta/g_1\sim 15$ we have $|d,1,1,1\rangle\to -|d,1,1,1\rangle$, $|d,1,1,0\rangle\to|a,1,1,0\rangle$, and $|d,1,0,0\rangle\to|d,1,0,0\rangle$ to a very good approximation. These conditions require $g_1\tau=3.5\times 10^4\pi$ and $g_2\tau=3.7\times 10^3\pi$.

III. CONCLUDING REMARK

In conclusion, we propose a scheme to realize a cavity QED based three-qubit quantum phase gate which may simplify the implementation of certain quantum computing problems. Our scheme contains two basic components: A four-level cascade atom and a three-mode cavity, where the photons in the three modes represent the three qubits. The realization of the phase gate requires appropriate detunings, coupling coefficients, and interaction times indicated in Eqs. (33) and (34). In general, the system must have a strong atom-field interaction and have a lifetime longer than the required interaction time.

The four-level cascade atomic structure is realistic and has been investigated a lot in the past [17–19]. For example, two-photon resonant four-wave mixing has been realized experimentally in a cascade four-level barium atom system [18,19], where the four energy levels a,b,c,d are formed by the ground state $6s^2\ {}^1S_0(|d\rangle)$, the intermediate state $6s6p\ {}^1P_1(|c\rangle)$, the Rydberg state $6s19d\ {}^1D_2(|b\rangle)$, and the doubly excited autoionizing Rydberg state $6p19d\ {}^{3/2}P(|a\rangle)$ [18,19]. The energy spacing between them is $\omega_{cd}=18\,060.3\ \mathrm{cm}^{-1}$, $\omega_{bc}=23\,562.1\ \mathrm{cm}^{-1}$, and $\omega_{ab}=21\,949.5\ \mathrm{cm}^{-1}$ [20,21]. From the available experimental data, the lifetime of $|c\rangle$ state is $\tau_1=8.3$ ns [22], while the lifetime of $|b\rangle$ state is $\tau_2=0.72\ \mu\mathrm{s}\gg\tau_1$ [23]. Since the field-atomic coupling constant $g\propto\tau^{-1/2}$, this system satisfies our requirement that $g_1\gg g_2$. It is worth pointing out that the state $|a\rangle$ is a two-electron resonance excited state. It can be obtained through a technology called isolated core excitation (ICE) [24]. The photons in modes 1 and 2 excite one of the electrons to the bound $6snd$ Rydberg state in which the electron is usually far from the ionic core. The third mode photon then excites the $6s$-$6p$ transition of the Ba^+ ion to reach the single autoionizing state $|a\rangle$. Obviously, such a cascade four-level energy structure is not limited to the barium atom. With the progress of nanoscience and semiconductor engineering, we may also be able to construct such a structure with certain artificial structures like quantum dots, quantum wells, or Josephson junctions.

As for the three-mode optical cavity, it has been widely used in laser physics and industry along with the development of multiwavelength lasers [25–27]. However, our scheme requires that the cavity works in the regime of "strong coupling" [28], which usually requires the optical modes to be confined in a small mode volume for extended periods of time. In other words, the cavity must have an extremely high Q factor. Various cavity techniques have been developed to realize strong coupling by employing high-finesse Fabry-Pérot optical microcavities [29], silica microsphere whispering gallery cavities [30,31], toroid microcavities [32], and photonic band-gap devices [33], where the Q factors with values exceeding 10^8 are achievable over a broad range of cavity diameters and wavelengths. Especially, ultrahigh Q factors approaching 10^{10} have been experimentally demonstrated [30], and Gotzinger *et al.* successfully achieved the strong coupling between multiple whispering-gallery modes and two individual nanoemitters (which have different center emission or absorption frequency) in one silicon microsphere resonator cavity [31]. With this kind of development in the resonator systems, it is promising that, in the near future, the strong interaction between a multimode field and a multilevel atom simultaneously inside one cavity can be experimentally realized.

Finally, it is worth noting that our proposal can be easily adapted to a microwave-ion or Rydberg atom system or a microwave-artificial atom system based on superconducting circuit quantum electrodynamics [34].

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APPENDIX

In the dressed state picture analysis, we ignored the possibility of excitation to the dressed state $|+\rangle$. In addition, when the initial state of the cavity field is $|1,1,0\rangle$, $|1,0,0\rangle$, or $|1,0,1\rangle$, there is a finite possibility for the atom exciting to upper levels. This is an important source of error. In the following part, we will derive the exact solutions for the probability amplitudes for each case.

1. Initial state: $|\psi_1\rangle=|d, 1, 0, 0\rangle$

For the initial state of $|d,1,0,0\rangle$, the wave vector at any time t is a superposition of $|d,1,0,0\rangle$ and $|c,0,0,0\rangle$, i.e., $|\psi_1\rangle=C_d(t)|d,1,0,0,0\rangle+C_c|c,0,0,0\rangle$. In the interaction picture, the probability amplitudes C_d and C_c satisfy the following Schrödinger equations:

$$\imath\hbar\frac{\partial C_d}{\partial t}=\hbar g_1 C_c e^{\imath\Delta t}, \tag{A1}$$

$$\imath\hbar\frac{\partial C_c}{\partial t}=\hbar g_1 C_d e^{-\imath\Delta t}. \tag{A2}$$

The initial conditions are $C_d(0)=1$ and $C_c(0)=0$. A solution of these equations subject to these conditions is

$$C_d(t) = \frac{1}{2}e^{\imath\Delta t/2}\left[\left(1 - \frac{\Delta}{\sqrt{\Delta^2+4g_1^2}}\right)e^{\imath\sqrt{\Delta^2+4g_1^2}t/2} + \left(1 + \frac{\Delta}{\sqrt{\Delta^2+4g_1^2}}\right)e^{-\imath\sqrt{\Delta^2+4g_1^2}t/2}\right], \quad (A3)$$

$$C_c(t) = -\frac{g_1}{\sqrt{\Delta^2+4g_1^2}}e^{-\imath\Delta t}\left(e^{\imath\sqrt{\Delta^2+4g_1^2}t/2} - e^{-\imath\sqrt{\Delta^2+4g_1^2}t/2}\right). \quad (A4)$$

2. Initial state: $|\psi_2\rangle = |d,1,0,1\rangle$

This case is the same as case 1. In the absence of the photon in mode of frequency ν_2, the photon in mode of frequency ν_3 would not interact with the atom.

3. Initial state: $|\psi_3\rangle = |d,1,1,0\rangle$

For the initial state $|d,1,1,0\rangle$, the wave function at any time is of the form $|\psi_t(t)\rangle = C_d(t)|d,1,1,0\rangle + C_c(t)|c,0,1,0\rangle + C_b(t)|b,0,0,0\rangle$. The corresponding equations for the amplitudes $C_d(t)$, $C_c(t)$, $C_b(t)$ are

$$\imath\hbar\frac{\partial C_d}{\partial t} = \hbar g_1 C_c e^{\imath\Delta t}, \quad (A5)$$

$$\imath\hbar\frac{\partial C_c}{\partial t} = \hbar g_1 C_d e^{-\imath\Delta t} + \hbar g_2 C_b e^{-\imath(\Delta+\delta)t}, \quad (A6)$$

$$\imath\hbar\frac{\partial C_b}{\partial t} = \hbar g_2 C_c e^{\imath(\Delta+\delta)t}. \quad (A7)$$

Eliminating C_d and C_b, we can get the following third-order time differential equation of C_c:

$$\frac{\partial^3 C_c}{\partial t^3} + \imath(2\Delta+\delta)\frac{\partial^2 C_c}{\partial t^2} + [g_1^2 + g_2^2 - \Delta(\Delta+\delta)]\frac{\partial C_c}{\partial t} + \imath[\Delta(g_1^2+g_2^2) + \delta g_1^2]C_c = 0. \quad (A8)$$

The initial conditions for $C_c(t=0)$ are

$$C_c(0) = 0, \quad \frac{\partial C_c(0)}{\partial t} = -\imath g_1, \quad \frac{\partial^2 C_c(0)}{\partial t^2} = -g_1\Delta. \quad (A9)$$

We consider a solution of the form

$$C_c(t) = Ae^{\omega_1 t} + Be^{\omega_2 t} + Ce^{\omega_3 t}, \quad (A10)$$

where ω_1, ω_2, and ω_3 are the roots of the third-order polynomial

$$z^3 + \imath(2\Delta+\delta)z^2 + [g_1^2 + g_2^2 + \Delta(\Delta+\delta)]z + \imath[\Delta(g_1^2+g_2^2) + g_1^2\delta]z = 0. \quad (A11)$$

The coefficients A, B, and C satisfy the following linear equations:

$$A + B + C = 0, \quad (A12)$$

$$A\omega_1 + B\omega_2 + C\omega_3 = -\imath g_1, \quad (A13)$$

$$A\omega_1^2 + B\omega_2^2 + C\omega_3^2 = -\imath g_1\Delta, \quad (A14)$$

yielding

$$A = \frac{\Delta - \imath(\omega_2+\omega_3)}{(\omega_1-\omega_2)(\omega_3-\omega_1)}g_1, \quad (A15)$$

$$B = \frac{\Delta - \imath(\omega_3+\omega_1)}{(\omega_2-\omega_3)(\omega_1-\omega_2)}g_1, \quad (A16)$$

$$C = \frac{\Delta - \imath(\omega_1+\omega_2)}{(\omega_3-\omega_1)(\omega_2-\omega_3)}g_1. \quad (A17)$$

Then we get C_d as follows:

$$C_d(t) = 1 - \frac{-\imath A}{\omega_1+\imath\delta} - \frac{-\imath B}{\omega_2+\imath\delta} - \frac{-\imath C}{\omega_3+\imath\delta} + e^{\imath\delta t}\left(\frac{-\imath A}{\omega_1+\imath\delta}e^{\omega_1 t} + \frac{-\imath B}{\omega_2+\imath\delta}e^{\omega_2 t} + \frac{-\imath C}{\omega_3+\imath\delta}e^{\omega_3 t}\right). \quad (A18)$$

4. Initial state: $|\psi_4\rangle = |d,1,1,1\rangle$

As before, we consider a general state of

$$|\psi_t(t)\rangle = C_d(t)|d,1,1,1\rangle + C_c(t)|c,0,1,1\rangle + C_b(t)|b,0,0,1\rangle + C_a(t)|a,0,0,0\rangle.$$

Using the Schrödinger equation in the interaction picture, we can get a group of first-order time differential equations of $C_d(t), C_c(t), C_b(t), C_a(t)$:

$$\imath\hbar\frac{\partial C_d}{\partial t} = \hbar g_1 C_c e^{\imath\Delta t}, \quad (A19)$$

$$\imath\hbar\frac{\partial C_c}{\partial t} = \hbar g_1 C_d e^{-\imath\Delta t} + \hbar g_2 C_b e^{-\imath(\Delta+\delta)t}, \quad (A20)$$

$$\imath\hbar\frac{\partial C_b}{\partial t} = \hbar g_2 C_c e^{\imath(\Delta+\delta)t} + \hbar g_3 C_a, \quad (A21)$$

$$\imath\hbar\frac{\partial C_a}{\partial t} = \hbar g_3 C_b. \quad (A22)$$

On eliminating C_c, C_b, C_d, we obtain the following fourth-order time differential equations for C_a:

$$\frac{\partial^4 C_a}{\partial t^4} - \imath(\Delta+2\delta)\frac{\partial^3 C_a}{\partial t^3} + [g_1^2 + g_2^2 + g_3^2 - \delta(\Delta+\delta)]\frac{\partial^2 C_a}{\partial t^2} - \imath[(\Delta+2\delta)g_3^2 + \delta g_2^2]\frac{\partial C_a}{\partial t} + [g_1^2 - \delta(\Delta+\delta)]g_3^2 C_a = 0. \quad (A23)$$

The initial conditions for C_a at $t=0$ are

$$C_a(0) = 0, \quad \frac{\partial C_a(0)}{\partial t} = 0, \quad \frac{\partial^2 C_a(0)}{\partial t^2} = 0,$$

$$\frac{\partial^3 C_a(0)}{\partial t^3} = \imath g_1 g_2 g_3. \tag{A24}$$

We consider a solution of the form

$$C_a(t) = Ae^{\omega_1 t} + Be^{\omega_2 t} + Ce^{\omega_3 t} + De^{\omega_4 t}, \tag{A25}$$

where ω_1, ω_2, ω_3, and ω_4 are the roots of the fourth-order polynomial equation

$$z^4 - \imath(\Delta + 2\delta)z^3 + [g_1^2 + g_2^2 + g_3^2 - \delta(\Delta + \delta)]z^2 - \imath[(\Delta + 2\delta)g_3^2 + \delta g_2^2]z + [g_1^2 - \delta(\Delta + \delta)]g_3^2 = 0. \tag{A26}$$

The coefficients A, B, C, and D satisfy the following linear equations:

$$A + B + C + D = 0, \tag{A27}$$

$$A\omega_1 + B\omega_2 + C\omega_3 + D\omega_4 = 0, \tag{A28}$$

$$A\omega_1^2 + B\omega_2^2 + C\omega_3^2 + D\omega_4^2 = 0, \tag{A29}$$

$$A\omega_1^3 + B\omega_2^3 + C\omega_3^3 + D\omega_4^3 = \imath g_1 g_2 g_3, \tag{A30}$$

yielding

$$A = \frac{\imath g_1 g_2 g_3}{(\omega_1 - \omega_2)(\omega_1 - \omega_3)(\omega_1 - \omega_4)}, \tag{A31}$$

$$B = \frac{\imath g_1 g_2 g_3}{(\omega_1 - \omega_2)(\omega_2 - \omega_3)(\omega_2 - \omega_4)}, \tag{A32}$$

$$C = \frac{\imath g_1 g_2 g_3}{(\omega_1 - \omega_3)(\omega_2 - \omega_3)(\omega_3 - \omega_4)}, \tag{A33}$$

$$D = \frac{\imath g_1 g_2 g_3}{(\omega_1 - \omega_4)(\omega_2 - \omega_4)(\omega_3 - \omega_4)}. \tag{A34}$$

From C_a, we can get C_d as follows:

$$C_d(t) = 1 + \frac{\imath g_1}{g_2 g_3}\left(\frac{A(\omega_1^2 + g_3^2)}{\omega_1 - \imath\delta}(e^{\omega_1 t - \imath\delta t} - 1) + \frac{B(\omega_2^2 + g_3^2)}{\omega_2 - \imath\delta}(e^{\omega_2 t - \imath\delta t} - 1) + \frac{C(\omega_3^2 + g_3^2)}{\omega_3 - \imath\delta}(e^{\omega_3 t - \imath\delta t} - 1) + \frac{D(\omega_4^2 + g_3^2)}{\omega_4 - \imath\delta}(e^{\omega_4 t - \imath\delta t} - 1)\right). \tag{A35}$$

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