Large two-atom two-photon vacuum Rabi oscillations in a high-quality cavity

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(Received 7 April 2004; published 11 October 2004)

We predict a large cooperative effect involving two-atom two-photon vacuum Rabi oscillations in a high-quality cavity. The two-photon emission occurs as a result of simultaneous deexcitation of both atoms with two-photon resonance condition $\omega_1+\omega_2\approx\omega_a+\omega_b$, where ω_1,ω_2 are the atomic transition frequencies and ω_a,ω_b are the frequencies of the emitted photons. The actual resonance condition depends on the vacuum Rabi couplings. The effect can be realized either with identical atoms in a bimodal cavity or with nonidentical atoms in a single-mode cavity.

DOI: 10.1103/PhysRevA.70.043807 PACS number(s): 42.50.Fx, 42.50.Hz, 42.50.Pq

I. INTRODUCTION

High-quality cavities have led to the study of a new regime of radiation matter interaction viz. the study of strongly interacting systems. Several new phenomena such as vacuum Rabi splittings [1–4], collapse and revival of Rabi oscillations [5], trapping states [6], and systems such as micromasers [7,8] have been studied. More recent applications on high-quality cavities are in the context of quantum computation [9]. Most of these works concern the interaction of the individual atoms. Earlier cooperative effects such as optical bistability involving a large number of atoms have been investigated [10]. A large part of these studies concerns the situations where the atomic transition frequency is almost equal to the cavity frequency. In this paper we report an unusual cooperative effect involving two atoms in a nonresonant cavity. This cooperative effect arises from the simultaneous deexcitation of two atoms such that the sum of the energies of emitted photons is equal to the sum of the excitation energies of the atoms. We demonstrate that in a high-quality cavity the two-atom two-photon resonant effect could be large, thus opening up the possibility of a variety of nonlinear, i.e., multiphoton cooperative phenomena in nonresonant cavities. For this purpose the recent development on the trapping of an atom inside the cavity [11] should be especially useful. We bring out the origin of such large two-atom two-photon Rabi oscillations.

We start by noting that in a two-photon emission process the two-photon resonance between the excited state $|e\rangle$ and the ground state $|g\rangle$ would occur at a frequency given by $\omega_{eg}=2\omega$, where ω_{eg} is the atomic transition frequency and ω is the frequency of the photons emitted. The process proceeds via intermediate states $|i\rangle$, which are away from a single-photon resonance. Now consider an interatomic process involving two atoms with distinct transition frequencies ω_1 and ω_2 such that $\omega_1-\omega$ and $\omega_2-\omega$ are large so that individual emissions are not important. However, as shown in Fig. 1(a), one can consider a two-photon emission process such that $\omega_1+\omega_2=2\omega$. Clearly this would be a cooperative process as it involves two atoms. In addition, it should also be important as it is a resonant process. Let us then examine the transition probability for such two-photon emission. Let H_+ be the interaction responsible for the emission of a photon defined by the interaction Hamiltonian which is written in the form

$$H_I = H_+ e^{i\omega t} + H_- e^{-i\omega t}. \tag{1}$$

Then second-order perturbation theory leads to the following expression for the rate of two-photon emission:

$$R_c = \frac{2\pi}{\hbar^2}\left| \frac{\langle g_1,g_2|H_+|g_1,e_2\rangle\langle g_1,e_2|H_+|e_1,e_2\rangle}{\hbar(\omega_1-\omega)} + \frac{\langle g_1,g_2|H_+|e_1,g_2\rangle\langle e_1,g_2|H_+|e_1,e_2\rangle}{\hbar(\omega_2-\omega)} \right|^2 \delta(\omega_1+\omega_2-2\omega). \tag{2}$$

Note that surprisingly $R_c=0$, as the two-photon matrix element vanishes when $\omega_1+\omega_2=2\omega$ as there are two paths for two-photon emission which interfere destructively. It has been argued that nonzero two-photon emission can result if we include interatomic interactions [12,13] which, however, are important only if the interatomic separation is less than a wavelength. A remarkable demonstration of such two-photon cooperative effects is given in a recent work [14] using the methods of single-molecule spectroscopy. Similar results apply to the case of two-photon emission by identical atoms [Fig. 1(b)] if the photons of frequencies ω_a and ω_b are emitted

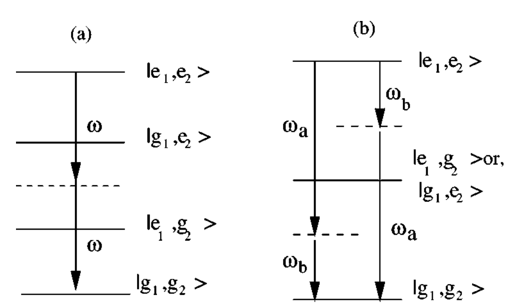

FIG. 1. Two ways for two-atom two-photon emission: (a) corresponding to two possible intermediate states $|e_1,g_2\rangle$ and $|g_1,e_2\rangle$ in the system of nonidentical atoms interacting with a single-mode vacuum and (b) in the system of identical atoms interacting with two modes of the vacuum.

$$\omega_a + \omega_b = 2\omega_0. \tag{3}$$

In this paper we examine such two-photon emission processes in a cavity. It is advantageous to use a cavity for the study of such a fundamental process as one would not be constrained by the requirement of small interatomic separation. We demonstrate how high-quality cavities can lead to a large two-photon Rabi oscillation involving two atoms. Note that vacuum Rabi oscillations in the context of a single atom interacting strongly with vacuum inside a single-mode resonant cavity have been studied extensively [1–4]. We also note that the two-photon micromaser in a single-mode cavity has been realized [8]. In this paper we consider two different cases of two-photon vacuum Rabi oscillations: (i) two identical atoms interacting with vacuum in a two-mode cavity and (ii) two nonidentical atoms in a single-mode cavity.

The paper is organized as follows. In Sec. II we consider the case of two identical atoms interacting with two modes of a cavity and discuss two-photon vacuum Rabi oscillations when the photons are emitted in different modes under resonance condition. In Sec. III we consider the case of two nonidentical atoms interacting with a single mode of the cavity. We present both approximate and analytical results. In Sec. IV, we confirm that the two-photon vacuum Rabi oscillations survive in the limit of small damping in a high-quality cavity. Finally in Sec. V, we present conclusions and future outlook.

II. TWO IDENTICAL ATOMS INTERACTING WITH VACUUM IN A BIMODAL CAVITY

We consider two identical two-level atoms, with transition frequency ω_0, interacting with two modes of the vacuum having frequencies ω_a and ω_b in a cavity as shown in Fig. 1(b). The Hamiltonian for the system is

$$H = \hbar\omega_a a^\dagger a + \hbar\omega_b b^\dagger b + \sum_{i=1,2}\hbar\left[\frac{\omega_0}{2}(|e_i\rangle\langle e_i| - |g_i\rangle\langle g_i|) + |e_i\rangle\langle g_i| \times (g_1 a + g_2 b) + |g_i\rangle\langle e_i|(g_1 a^\dagger + g_2 b^\dagger)\right], \tag{4}$$

where a and $a^\dagger$ (b and $b^\dagger$) are annihilation and creation operators for the first (second) mode of the cavity and g_1 and g_2 are the coupling constants. In a frame rotating with frequency ω_0, the Hamiltonian (4) becomes

$$H = -\hbar\Delta a^\dagger a - \hbar\delta b^\dagger b + \sum_{i=1,2}\hbar[|e_i\rangle\langle g_i|(g_1 a + g_2 b) + |g_i\rangle\langle e_i| \times (g_1 a^\dagger + g_2 b^\dagger)],$$

$$\Delta = \omega_0 - \omega_a, \quad \delta = \omega_0 - \omega_b. \tag{5}$$

We consider the special case of two-photon emission, i.e., the case when the initial state of the atom-cavity system is

$$|\psi(0)\rangle = |e_1, e_2, 0, 0\rangle. \tag{6}$$

Considering all possible states of the system in evolution, the state of the system at time t can be written as

$$|\psi(t)\rangle = c_1(t)|e_1,e_2,0,0\rangle + \frac{1}{\sqrt{2}}(|e_1,g_2\rangle + |g_1,e_2\rangle)\{c_2(t)|1,0\rangle + c_3(t)|0,1\rangle\} + |g_1,g_2\rangle\{c_4(t)|1,1\rangle + c_5(t)|2,0\rangle + c_6(t)|0,2\rangle\}. \tag{7}$$

Different terms in the wave function (7) correspond to no-photon emission, one-photon emission, and two-photon emission. The photon emission can take place in either mode. A very interesting aspect of the state (7) is its entangled nature. This provides a method of producing entangled states, say, entanglement of two-cavity modes [15]. The time-dependent amplitudes $c_i(t)$ are determined by

$$\dot{c}_1 = -ig_1\sqrt{2}c_2 - ig_2\sqrt{2}c_3,$$

$$\dot{c}_2 = i\Delta c_2 - ig_1\sqrt{2}c_1 - ig_2\sqrt{2}c_4 - 2ig_1c_5,$$

$$\dot{c}_3 = i\delta c_3 - ig_2\sqrt{2}c_1 - ig_1\sqrt{2}c_4 - 2ig_2c_6,$$

$$\dot{c}_4 = i(\Delta+\delta)c_4 - ig_2\sqrt{2}c_2 - ig_1\sqrt{2}c_3,$$

$$\dot{c}_5 = 2i\Delta c_5 - 2ig_1c_2,$$

$$\dot{c}_6 = 2i\delta c_6 - 2ig_2c_3. \tag{8}$$

The complete solution of Eq. (8) has 6 eigenvalues corresponding to those where there will be a 15 peak spectrum. In order to understand the nature of the two-atom two-photon resonance we present numerical as well as approximate analysis which can capture the physics of the cooperative process. We consider the case when detunings to the cavity field are much larger than the couplings, i.e., $|\Delta|, |\delta| \gg g_1, g_2$ but $|\Delta+\delta|$ is small and the condition for two-photon resonance is $\Delta+\delta=0$. In such a case the cooperative two-photon process should dominate and single-photon processes would be insignificant. The results of numerical integration of Eq. (8) are plotted in Fig. 2. In the case when $g_1 \neq g_2$ a novel resonance is achieved. The probability of two-photon emission at resonance is quite high. The resonance is shifted from the position $\Delta+\delta=0$. This shift is due to the strong coupling to the vacuum field in the cavity. For $g_2/g_1=1.5$ and $\Delta=-5g_1$ maximum two-photon emission probability is approximately 0.9 and the interaction time required for achieving maximum probability is given by $g_1 t \approx 6\pi$.

Having established numerically that the two-photon resonance can be large in cavities, we present an approximate analysis to demonstrate it. Under the abovementioned conditions for two-photon resonance we can eliminate fast oscillating variables c_2, c_3, c_5, c_6 and effectively reduce the dynamics in terms of slowly oscillating variables c_1 and c_4. A simple treatment where one sets $\dot{c}_2=\dot{c}_3=\dot{c}_5=\dot{c}_6=0$ does not yield the physics of the two-atom two-photon emission. We thus relegate the procedure for eliminating fast variables to the Appendix. The reduced form of Eq. (8) is written as

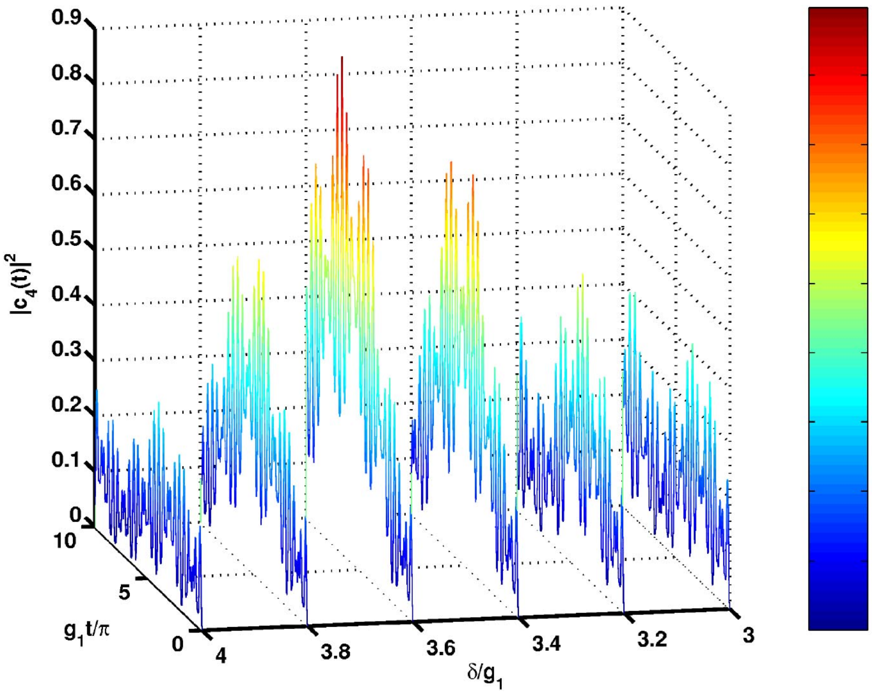

FIG. 2. (Color online) Two-atom two-photon emission probability $|c_4(t)|^2$ in a system of identical atoms interacting with vacuum in a two-mode cavity for $g_2/g_1=1.5$ and $\Delta/g_1=-5.0$.

$$\dot{c}_1 = -i\left(\frac{2g_1^2\Delta}{\Delta^2-2g_1^2}+\frac{2g_2^2\delta}{\delta^2-2g_2^2}\right)c_1 + 2ig_1g_2\left(\frac{\Delta}{\Delta^2+2g_1^2} + \frac{\delta}{\delta^2+2g_2^2}\right)c_4,$$

$$\dot{c}_4 = 2ig_1g_2\left(\frac{\Delta}{\Delta^2+2g_1^2}+\frac{\delta}{\delta^2+2g_2^2}\right)c_1 + i\left(\Delta+\delta-\frac{2g_1^2\delta}{\delta^2-2g_2^2} - \frac{2g_2^2\Delta}{\Delta^2-2g_1^2}\right)c_4. \tag{9}$$

The solution of Eq. (9) gives

$$|c_4(t)|^2 = \frac{4G^2}{4G^2+\Omega^2}\sin^2\frac{\sqrt{4G^2+\Omega^2}t}{2}, \tag{10}$$

with

$$G = 2g_1g_2\left(\frac{\Delta}{\Delta^2+2g_1^2}+\frac{\delta}{\delta^2+2g_2^2}\right),$$

$$\Omega = \Delta+\delta+2(g_1^2-g_2^2)\left(\frac{\Delta}{\Delta^2-2g_1^2}-\frac{\delta}{\delta^2-2g_2^2}\right). \tag{11}$$

Note that in the limit $g_1=g_2$ and $\Delta+\delta=0$, the probability amplitude c_4 for two-photon emission tends to zero, as both Ω and the numerator in Eq. (10) become proportional to $(\Delta+\delta)$. Thus when couplings to the modes are the same, the two-photon emission probability has no resonance. In this case the transitions from $|e_1,e_2,0,0\rangle$ to $|g_1,g_2,1,1\rangle$ via states $(1/\sqrt{2})(|e_1,g_2\rangle+|g_1,e_2\rangle)|1,0\rangle$ and $(1/\sqrt{2})(|g_1,e_2\rangle+|e_1,g_2\rangle)|0,1\rangle$ interfere destructively. We further note that to order $g_1^2g_2^2$ the two-photon resonance does not occur:

$$|c_4(t)|^2 = \frac{16g_1^2g_2^2}{\delta^2\Delta^2}\sin^2\frac{\delta t}{2}\sin^2\frac{\Delta t}{2}. \tag{12}$$

The usual second-order perturbation theory cannot lead to interatomic two-photon resonance. One has to consider higher-order terms in g_1 and g_2. However, then the excitation itself would be negligible. Therefore one needs high-quality cavities. The probability of cooperative emission of two-photons in different modes is a periodic function of time. In Fig. 3(a), we plot the maximum value of $|c_4(t)|^2$ as a function of δ and in Fig. 3(b) as a function of time t, for fixed values of g_1, g_2, and Δ. At two-photon resonance the probability corresponding to two-photon emission in one of the two modes is much smaller than the probability of two-photon emission in different modes. From Eqs. (10) and (11) it is clear that the two-photon resonance occurs at $\Delta+\delta+4(g_1^2/\Delta+g_2^2/\delta)\approx 0$. Thus the interaction with the cavity modifies the condition of two-photon resonance. This is seen quite clearly in Fig. 3(a). We note the connection of the resonance frequency Ω to the one-photon Stark shifts. It is well known that the shift in the frequency of a two-level atom in the presence of a field with n photons is given by $2g^2(n+1)/\Delta$ which is equal to $4g^2/\Delta$ for $n=1$. Thus the change $4(g_1^2/\Delta+g_2^2/\delta)$ is equal to the frequency shift of both the atoms due to the presence of a single photon. We have checked using the full solution of the Schrodinger equation that the result (10) is quite good. However, it should be borne in mind that the exact result is not periodic and exhibits rapid variations though the envelope agreeing with the result (10). The abovementioned approximate results are valid for larger values of detunings but for larger values of detunings a large interaction time is required to reach the maximum of two-atom two-photon transition probability. This should be possible with the recently developed method of trapping atoms

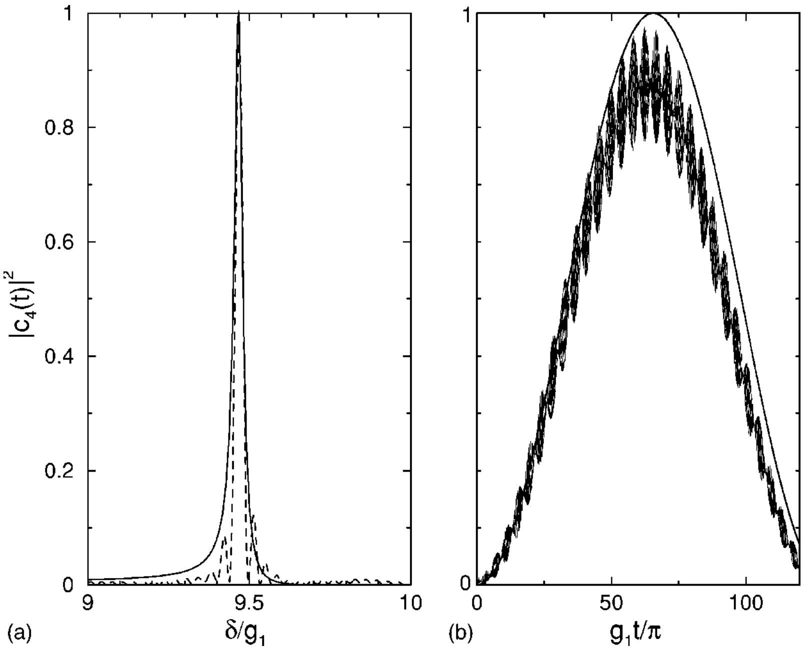

FIG. 3. The maximum value of the two-atom two-photon emission probability $|c_4(t)|^2$ in the system of two identical atoms interacting with vacuum in a two-mode cavity is plotted with respect to (a) detuning δ and (b) time for $g_2/g_1 = 1.5$ and $\Delta = -10g_1$. The solid line corresponds to the approximate result and the dotted line corresponds to the exact numerical result.

in a cavity [11]. The other possibility is to work under the conditions of Fig. 2.

III. TWO-PHOTON EMISSION BY TWO NONIDENTICAL ATOMS IN A SINGLE-MODE CAVITY

In this section we analyze a system of two nonidentical atoms interacting with a single-mode vacuum field in a cavity [Fig. 1(a)]. Consider two nonidentical two-level atoms with excited states $|e_1\rangle, |e_2\rangle$ and their ground states $|g_1\rangle, |g_2\rangle$ interacting with a single-mode cavity field of frequency ω. The Hamiltonian of this system is

$$H = \hbar\left[\frac{\omega_1}{2}(|e_1\rangle\langle e_1| - |g_1\rangle\langle g_1|) + \frac{\omega_2}{2}(|e_2\rangle\langle e_2| - |g_2\rangle\langle g_2|) + \omega a^\dagger a\right] + \hbar g_1(|e_1\rangle\langle g_1|a + a^\dagger|g_1\rangle\langle e_1|) + \hbar g_2(|e_2\rangle\langle g_2|a + a^\dagger|g_2\rangle\langle e_2|), \tag{13}$$

where $\omega_1(\omega_2)$ is transition frequency for the first (second) atom, a and $a^\dagger$ are annihilation and creation operators for the field, and $g_1(g_2)$ is the coupling constant to the cavity mode with the first (second) atom. In a rotating frame the Hamiltonian H can be written as

$$H = -\hbar\Delta|g_1\rangle\langle g_1| - \hbar\delta|g_2\rangle\langle g_2| + \hbar g_1(|e_1\rangle\langle g_1|a + a^\dagger|g_1\rangle\langle e_1|) + \hbar g_2(|e_2\rangle\langle g_2|a + a^\dagger|g_2\rangle\langle e_2|), \tag{14}$$

$$\Delta = \omega_1 - \omega, \quad \delta = \omega_2 - \omega. \tag{15}$$

Let us consider an initial state $|\psi(0)\rangle = |e_1, e_2, 0\rangle$ with both atoms in the excited state and cavity in the vacuum state. The state of the system at time t can be written as

$$|\psi(t)\rangle = c_1(t)|e_1,e_2,0\rangle + c_2(t)|e_1,g_2,1\rangle + c_3(t)|g_1,e_2,1\rangle + c_4(t)|g_1,g_2,2\rangle, \tag{16}$$

where the expansion coefficients c's satisfy

$$\dot{c}_1 = -ig_2c_2 - ig_1c_3,$$

$$\dot{c}_2 = i\delta c_2 - ig_1\sqrt{2}c_4 - ig_2c_1,$$

$$\dot{c}_3 = i\Delta c_3 - ig_2\sqrt{2}c_4 - ig_1c_1,$$

$$\dot{c}_4 = i(\Delta + \delta)c_4 - ig_1\sqrt{2}c_2 - ig_2\sqrt{2}c_3. \tag{17}$$

The two-photon resonance condition for this system would be $\Delta + \delta = 0$. For couplings g_1, g_2 much smaller than $|\Delta|, |\delta|$, the solution of Eq. (17) gives

$$c_4(t) = -\frac{4g_1g_2\sqrt{2}}{\delta\Delta}\sin\frac{\delta t}{2}\sin\frac{\Delta t}{2} + (\text{higher-order terms}). \tag{18}$$

The first term in Eq. (18) represents independent emission by each atom. Clearly, to lowest order in g_1g_2 no two-photon resonance occurs. Such a resonance can come from the terms of higher order. Assuming that $|\Delta|$ and $|\delta|$ are large but $|\Delta + \delta|$ is small, we eliminate fast oscillating variables c_2 and c_3 in a way similar to the previous case and Eq. (17), in terms of slowly oscillating variables reduces, to

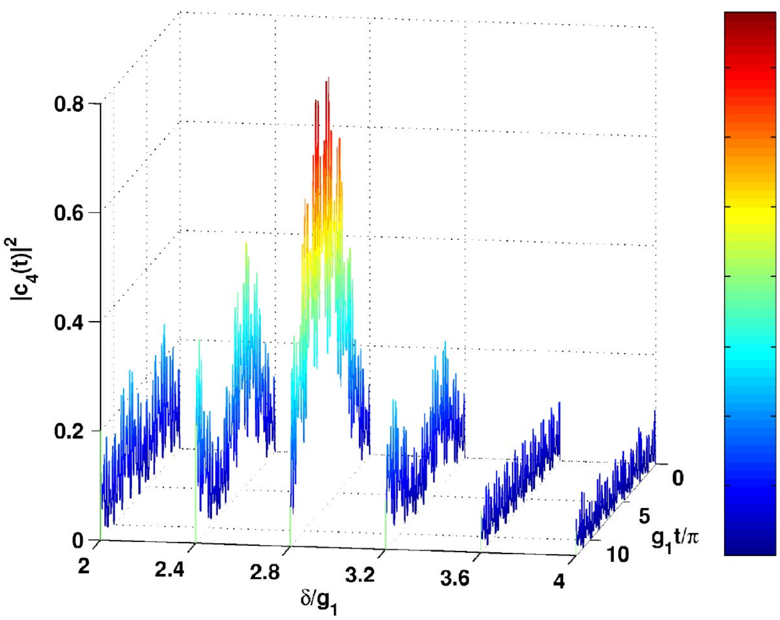

FIG. 4. (Color online) Two-atom two-photon emission probability $|c_4(t)|^2$ in a system of non-identical atoms interacting with vacuum in a single-mode cavity for $\Delta/g_1=-5.0$ and $g_2/g_1=2.0$.

$$\dot{c}_1 = -i\left(\frac{g_1^2}{\Delta} + \frac{g_2^2}{\delta}\right)c_1 + ig_1g_2\sqrt{2}\left(\frac{\Delta}{\Delta+2g_1^2} + \frac{\delta}{\delta+2g_2^2}\right)c_4,$$

$$\dot{c}_4 = ig_1g_2\sqrt{2}\left(\frac{\Delta}{\Delta+2g_1^2} + \frac{\delta}{\delta+2g_2^2}\right)c_1 + i\left(\Delta+\delta+\frac{2g_1^2}{\Delta}+\frac{2g_2^2}{\delta}\right)c_4. \tag{19}$$

We find the approximate result for the two-photon emission probability

$$|c_4(t)|^2 = \frac{4G'^2}{4G'^2+\Omega'^2}\sin^2\frac{\sqrt{4G'^2+\Omega'^2}t}{2}, \tag{20}$$

with

$$G' = \sqrt{2}g_1g_2\left(\frac{\Delta}{\Delta^2+2g_1^2} + \frac{\delta}{\delta^2+2g_2^2}\right),$$

$$\Omega' = \Delta+\delta+3\left(\frac{g_1^2}{\Delta}+\frac{g_2^2}{\delta}\right). \tag{21}$$

For large $|\Delta|$ and $|\delta|$, Eq. (20) shows two-photon resonance at $\Delta+\delta+3(g_1^2/\Delta+g_2^2/\delta)\approx 0$. Further such two-atom two-photon resonances appear for $g_1\neq g_2$, which disappears when $g_1=g_2$. In the latter case the antisymmetric state $(|g_1,e_2,1\rangle-|e_1,g_2,1\rangle)/\sqrt{2}$ is decoupled from $|e_1,e_2,0\rangle$ and $|g_1,g_2,2\rangle$. We present numerical results in Fig. 4. The graph shows two-photon resonance for $g_1\neq g_2$. It is clear that the position of resonance is shifted from $\Delta+\delta=0$. This shift in the position of resonance is due to larger values of g_1 and g_2 and depends on the ratio g_2/g_1. There is a large enhancement in the probability of two-photon resonant emission in a high-quality cavity. It is expected that such effects can be studied by placing the system used by Hettich *et al.* [14] in a cavity.

IV. EFFECTS OF CAVITY DAMPING

Before concluding we examine the effect of cavity decay on two-atom two-photon vacuum Rabi oscillations. We do a calculation based on a master equation. Let $2\kappa_a$ and $2\kappa_b$ be the rate of loss of photons from the first and second modes, respectively. The density matrix of the system of two atoms interacting with two mode field in the cavity will evolve according to the master equation

$$\dot{\rho} = -\frac{i}{\hbar}[H,\rho] - \kappa_a(a^\dagger a\rho - 2a\rho a^\dagger + \rho a^\dagger a) - \kappa_b(b^\dagger b\rho - 2b\rho b^\dagger + \rho b^\dagger b). \tag{22}$$

The density matrix for this system can be expressed in terms of all the states which are generated by the combined effect of H and dissipation. Because of the cavity decay, many more states are involved in the dynamics. For example, for identical atoms interacting in a bimodal cavity, the relevant states are $|e_1,e_2,0,0\rangle$, $|g_1,e_2,0,0\rangle$, $|g_1,e_2,1,0\rangle$, $|g_1,e_2,0,1\rangle$, $|e_1,g_2,0,0\rangle$, $|e_1,g_2,1,0\rangle$, $|e_1,g_2,0,1\rangle$, $|g_1,g_2,0,0\rangle$, $|g_1,g_2,0,1\rangle$, $|g_1,g_2,1,0\rangle$, $|g_1,g_2,0,2\rangle$, $|g_1,g_2,1,1\rangle$, and $|g_1,g_2,2,0\rangle$. For this system the density matrix is expressed as

$$\rho \equiv \sum_{i',j',i,j=0}^{1}\sum_{k'=0}^{i'+j'}\sum_{k=0}^{i+j}\sum_{l'=0}^{i'+j'-k'}\sum_{l=0}^{i+j-k}\rho(i',j',k',l',i,j,k,l) \times|i',j',k',l'\rangle\langle i,j,k,l|. \tag{23}$$

Here i,i' (j,j') represent states of the first (second) atom with the convention $|0\rangle$ corresponding to the excited state and $|1\rangle$ corresponding to the ground state, the indices k,k' (l,l') represent the number of photons in the first (second) mode. Thus the dissipation requires considerable numerical work. Results for two identical atoms in a bimodal cavity are shown in Fig. 5. We show results for optical cavities with $g/\kappa\approx 30$ in Fig. 5(b) and for currently realizable cavities $(g/\kappa=10)$ in Fig. 5(c). The two-atom two-photon vacuum

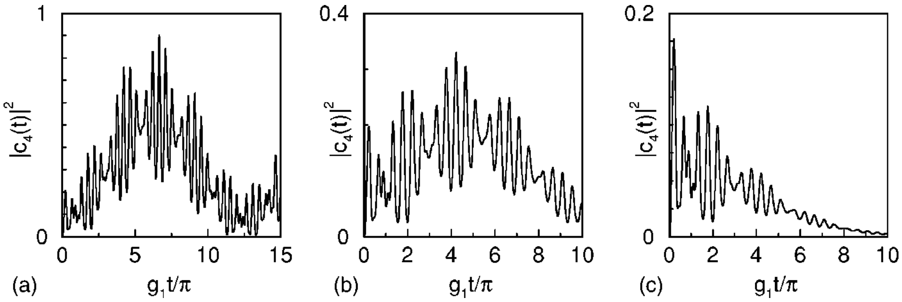

FIG. 5. Periodic behavior of two-atom two-photon emission probability $|c_4(t)|^2$ for identical atoms interacting with vacuum in a bimodal cavity for $\delta=3.5g_1$, $\Delta=-5g_1$, $g_2=1.5g_1$, and cavity damping constants (a) $\kappa_a=\kappa_b=0.00$, (b) $\kappa_a=\kappa_b=0.03g_1$, (c) $\kappa_a=\kappa_b=0.1g_1$.

Rabi oscillations survive in the limit of small damping $g/\kappa \approx 30$ but for larger damping $(g/\kappa=10)$ die fast. Similar results are found for two nonidentical atoms in a single-mode cavity.

V. CONCLUSIONS

We have reported large two-atom two-photon vacuum Rabi oscillations in two systems, one having two identical atoms in a two-mode cavity and another having two nonidentical atoms in a single-mode cavity. We have shown that for asymmetric couplings $(g_1 \neq g_2)$, the probability of two-photon emission is quite large but for symmetric couplings $(g_1=g_2)$, the two-photon emission probability is very small. Further, we have shown that the condition of two-photon resonance in the case of strong atom-field interaction is modified from its free-space form $(\Delta+\delta=0)$. These two-photon transitions involving two atoms can be used for generating and detecting different types of entanglement between two field modes and two atoms [16].

ACKNOWLEDGMENTS

G.S.A. thanks V. Sandoghdar and G. Rempe for discussions on the two-atom two-photon resonance.

APPENDIX

Our procedure for eliminating fast oscillating variables is extended form of the procedure discussed in Ref. [17]. The Hamiltonian (5) can be written as

$$H = H_0 + \epsilon V,$$

where

$$H_0 = -\Delta a^\dagger a - \delta b^\dagger b,$$

$$\epsilon V = \sum_{i=1,2} \hbar [|e_i\rangle\langle g_i|(g_1 a + g_2 b) + |g_i\rangle\langle e_i|(g_1 a^\dagger + g_2 b^\dagger)]. \tag{A1}$$

The eigenstates and corresponding eigenvalues of H_0 are

$$|1\rangle \equiv |e_1,e_2,0,0\rangle, \quad E_1 \equiv 0,$$

$$|2\rangle \equiv 2^{-1/2}(|e_1,g_2\rangle + |g_1,e_2\rangle)|1,0\rangle, \quad E_2 \equiv -\Delta,$$

$$|3\rangle \equiv 2^{-1/2}(|e_1,g_2\rangle + |g_1,e_2\rangle)|0,1\rangle, \quad E_3 \equiv -\delta,$$

$$|4\rangle \equiv |g_1,g_2,1,1\rangle, \quad E_4 \equiv -(\Delta+\delta),$$

$$|5\rangle \equiv |g_1,g_2,2,0\rangle, \quad E_5 \equiv -2\Delta,$$

$$|6\rangle \equiv |g_1,g_2,0,2\rangle, \quad E_6 \equiv -2\delta.$$

The resolvent for H_0 is the function

$$G_0(z) = \frac{1}{z - H_0}, \tag{A2}$$

where z is complex. If P_i is projection operator for the eigenstates of H_0

$$P_i = |i\rangle\langle i|, \quad i = 1,2, \ldots ,6. \tag{A3}$$

The resolvent G_0 can be expressed as

$$G_0(z) = \sum_i \frac{P_i}{z - E_i}. \tag{A4}$$

The resolvent for the full Hamiltonian H is

$$\begin{aligned} G(z) &= \frac{1}{z - H_0 - \epsilon V}, \\ &= \frac{1}{z - H_0}\left(1 + \epsilon V \frac{1}{z - H}\right), \\ &= G_0(1 + \epsilon V G). \end{aligned} \tag{A5}$$

From Eq. (A5) the resolvent for the full Hamiltonian H can be expressed in the power series of ϵ as

$$G = \sum_n \epsilon^n G_0 (V G_0)^n. \tag{A6}$$

For small values of ϵ, $G(z)$ has singularities in the complex z plane in the neighborhood of poles of function G_0, i.e., eigenvalues of H_0. Further eigenvalues E_1 and E_4 are very close to each other under the condition $\Delta+\delta \approx 0$ and other eigenvalues are largely separated. We consider a contour, Γ in the z plane that encloses eigenvalues E_1 and E_4 only and leaves others outside as shown in Fig. 6. We define a new projection operator P_Γ as

$$\begin{aligned} P_\Gamma &= \overline{P_1} + \overline{P_4}, \\ &= \frac{1}{2i\pi}\oint_\Gamma G(z)dz. \end{aligned} \tag{A7}$$

Here $\overline{P_1}$ and $\overline{P_4}$ are the projection operators for eigenstates of

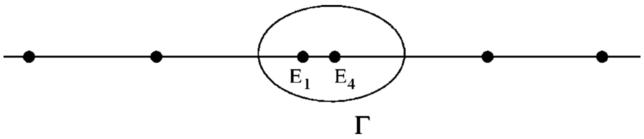

FIG. 6. The contour in the complex plane shielding two eigenvalues E_1 and E_4 and leaving others outside.

full Hamiltonian H corresponding to the eigenvalues inside the contour. The effective Hamiltonian will have the form

$$H_{\text{eff}} \equiv (P_1 + P_4) H P_\Gamma (P_1 + P_4). \tag{A8}$$

From the definition of the resolvent we have

$$(z - H)G \equiv G(z - H) \equiv 1,$$

$$HP_\Gamma = \frac{1}{2i\pi}\oint_\Gamma zG(z)dz. \tag{A9}$$

Substituting the value of $G(z)$ from Eq. (A6) into Eq. (A9) and interchanging summation to the integration we have

$$HP_\Gamma = \sum_n \frac{1}{2i\pi}\oint_\Gamma zG_0(VG_0)^n dz. \tag{A10}$$

The effective Hamiltonian can be expressed as

$$H_{\text{eff}} = E_1P_1 + E_4P_4 + \sum_{n=1}^{\infty} \epsilon^n A^{(n)},$$

$$A(n) = (P_1 + P_4)\sum_{n=1}^{\infty} \frac{1}{2i\pi}\oint_\Gamma zG_0(VG_0)^n dz (P_1 + P_4). \tag{A11}$$

Inside the contour Γ, G_0 has singularities at E_1 and E_4 only so the integral in Eq. (A11) is nothing but the sum of the residues at $z = E_1$ and $z = E_4$. Further as in our case ϵP_1VP_1, ϵP_4VP_4, and ϵP_1VP_4 equal to zero, there is no first-order and third-order terms. The second order term is

$$A^{(2)} = P_1VQ_1VP_1 + P_4VQ_4VP_4 + P_1VQ_4VP_4 + P_4VQ_4VP_1,$$

$$Q_j = \sum_{i\neq 1,4} \frac{P_i}{E_j - E_i}. \tag{A12}$$

The fourth-order term is

$$A^{(4)} = \frac{1}{2i\pi}\oint_\Gamma z\left(\frac{P_1}{z-E_1} + \frac{P_4}{z-E_4}\right)V\sum_{i\neq 1,4}\frac{P_i}{z-E_i}V\left(\frac{P_1}{z-E_1} + \frac{P_4}{z-E_4} + \sum_{j\neq 1,4}\frac{P_j}{z-E_j}\right)V\sum_{k\neq 1,4}\frac{P_k}{z-E_k}V\left(\frac{P_1}{z-E_1} + \frac{P_4}{z-E_4}\right)dz. \tag{A13}$$

For simplification we use the condition for resonance $\Delta + \delta = 0$, i.e., $E_1 = E_4$. Thus the fourth-order term is

$$A^{(4)} = \frac{1}{2i\pi}\oint_\Gamma z\left(\frac{P_1}{z-E_1} + \frac{P_4}{z-E_1}\right) \times V\sum_{i\neq 1,4}\frac{P}{z-E_1}V\sum_{j\neq 1,4}\frac{P_j}{z-E_j}V\sum_{k\neq 1,4}\frac{P_k}{z-E_k}V\left(\frac{P_1}{z-E_1} + \frac{P_4}{z-E_1}\right)dz. \tag{A14}$$

Integrating Eq. (A14) we have the fourth-order term

$$A^{(4)} = (P_1 + P_4)VQ_1VQ_1VQ_1V(P_1 + P_4). \tag{A15}$$

Using the values of E_1, E_2, E_3, E_4, E_5, E_6, and V the effective Hamiltonian expressed in basis $|e_1, e_2, 0, 0\rangle$ and $|g_1, g_2, 1, 1\rangle$ is

$$H_{\text{eff}} = \begin{bmatrix} \dfrac{2g_1^2}{\Delta} + \dfrac{2g_2^2}{\delta} + \dfrac{4g_1^4}{\Delta^3} + \dfrac{4g_2^4}{\delta^3} & -\dfrac{2g_1g_2}{\Delta} - \dfrac{2g_1g_2}{\delta} + \dfrac{4g_1^3g_2}{\Delta^3} + \dfrac{4g_1g_2^3}{\delta^3} \\ -\dfrac{2g_1g_2}{\Delta} - \dfrac{2g_1g_2}{\delta} + \dfrac{4g_1^3g_2}{\Delta^3} + \dfrac{4g_1g_2^3}{\delta^3} & -(\Delta + \delta) - \dfrac{2g_2^2}{\delta} - \dfrac{2g_1^2}{\Delta} + \dfrac{4g_1^2g_2^2}{\Delta^3} + \dfrac{4g_1^2g_2^2}{\delta^3} \end{bmatrix}. \tag{A16}$$

With some algebraic manipulation and considering g_1 and g_2 up to fourth order effectively the Hamiltonian (5) reduces to

$$H_{\text{eff}} = \begin{bmatrix} \dfrac{2g_1^2\Delta}{\Delta^2 - 2g_1^2} + \dfrac{2g_2^2\delta}{\delta^2 - 2g_2^2} & -2g_1g_2\left(\dfrac{\Delta}{\Delta^2 + 2g_1^2} + \dfrac{\delta}{\delta^2 + 2g_2^2}\right) \\ -2g_1g_2\left(\dfrac{\Delta}{\Delta^2 + 2g_1^2} + \dfrac{\delta}{\delta^2 + 2g_2^2}\right) & -\left(\Delta + \delta - \dfrac{2g_2^2\Delta}{\Delta^2 - 2g_1^2} - \dfrac{2g_1^2\delta}{\delta^2 - 2g_2^2}\right) \end{bmatrix}. \tag{A17}$$

It should be noted here that as two-atom two-photon resonance appears at large interaction time in dispersive limit, the terms in the effective Hamiltonian up to forth order are important to predict the correct evolution. Using the effective Hamiltonian (A17), Eq. (8) reduces to Eq. (9).

[1] J. J. Sanchez-Mondragon, N. B. Narozhny, and J. H. Eberly, Phys. Rev. Lett. **51**, 550 (1983).
[2] G. S. Agarwal, J. Opt. Soc. Am. B **2**, 480 (1985); G. S. Agarwal, Phys. Rev. Lett. **53**, 1732 (1984).
[3] H. J. Kimble, in *Cavity Quantum Electrodynamics*, edited by P. Berman (Academic Press, London, 1994), p. 203.
[4] T. W. Mossberg and M. Lewenstein, in *Cavity Quantum Electrodynamics* (Ref. [3]), p.171.
[5] J. H. Eberly, N. B. Narozhny, and J. J. Sanchez-Mondragon, Phys. Rev. Lett. **44**, 1323 (1980).
[6] M. Weidinger, B. T. H. Varcoe, R. Heerlein, and H. Walther, Phys. Rev. Lett. **82**, 3795 (1999); P. Meystre, G. Rempe, and H. Walther, Opt. Lett. **13**, 1078 (1988).
[7] G. Raithel, C. Wagner, H. Walther, L. M. Narducci, and M. O. Scully, in *Cavity Quantum Electrodynamics*, edited by P. Berman (Academic, London, 1994), p. 57.
[8] L. Davidovich, J. M. Raimond, M. Brune, and S. Haroche, Phys. Rev. A **36**, 3771 (1987); M. Brune, J. M. Raimond, and S. Haroche, *ibid.* **35**, 154 (1987); M. Brune, J. M. Raimond, P. Goy, L. Davidovich, and S. Haroche, Phys. Rev. Lett. **59**, 1899 (1987).
[9] J. M. Raimond, M. Brune, and S. Haroche, Rev. Mod. Phys. **73**, 565 (2001).
[10] L. A. Lugiato, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1984), Vol. XXI, p. 69.
[11] J. McKeever, J. R. Buck, A. D. Boozer, A. Kuzmich, H.-C. Nägerl, D. M. Stamper-Kurn, and H. J. Kimble, Phys. Rev. Lett. **90**, 133602 (2003).
[12] G. V. Varada and G. S. Agarwal, Phys. Rev. A **45**, 6721 (1992); D. L. Andrews and N. P. Blake, J. Mod. Opt. **37**, 701 (1990).
[13] A. Beige and G. C. Hegerfeldt, Phys. Rev. A **58**, 4133 (1998); E. V. Goldstein and P. Meystre, *ibid.* **56**, 5135 (1997).
[14] C. Hettich, C. Schmitt, J. Zitzmann, S. Kuhn, I. Gerhardt, and V. Sandoghdar, Science **298**, 385 (2002).
[15] A. Rauschenbeutel, P. Bertet, S. Osnaghi, G. Nogues, M. Brune, J. M. Raimond, and S. Haroche, Phys. Rev. A **64**, 050301 (2001).
[16] J. D. Franson and T. B. Pittman, Phys. Rev. A **60**, 917 (1999).
[17] A. Messiah, *Quantum Mechanics* (Dover, London, 1999), p. 685.