# Cavity-induced two-photon absorption in unidentical atoms

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We predict the existence of cavity-induced two-photon absorption in unidentical atoms, and demonstrate the nonclassical character of this two-photon absorption. We show the effects of photon statistics on cavity induced two-photon absorption. We also note the possibility of trapping states in the present system. [S1050-2947(98)06904-2]

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## I. INTRODUCTION

The transition probability for two-photon absorption, as calculated first by Goepport Mayor [1], depends on the value of the two-photon matrix element defined by

$$\wp \equiv \sum_j \frac{\langle f|\vec{d}\cdot\vec{\varepsilon}|j\rangle\langle j|\vec{d}\cdot\vec{\varepsilon}|i\rangle}{E_{ji}-\hbar\omega}, \tag{1}$$

where $\vec{\varepsilon}$ and $\omega$ are, respectively, the amplitude and frequency of the electromagnetic field, $E_{ji}$ is the transition energy between atomic energy levels $|i\rangle$ and $|j\rangle$, and $\vec{d}$ is the dipole moment operator. The two-photon absorption will vanish if the matrix element $\wp$ becomes zero. Many examples are known in the literature, where $\wp$ can be zero [2,3]. An example of a system which is particularly interesting consists of two unidentical two-level atoms $A$ and $B$ with transition frequencies $\omega_A$ and $\omega_B$. Let us denote by $|g\rangle$ and $|e\rangle$ the lower and upper states of the transition. It can be seen that we have two different paths for two-photon absorption:

$$\begin{aligned}&|g_A,g_B\rangle\rightarrow|e_A,g_B\rangle\rightarrow|e_A,e_B\rangle,\\ &|g_A,g_B\rangle\rightarrow|g_A,e_B\rangle\rightarrow|e_A,e_B\rangle.\end{aligned} \tag{2}$$

These two transition amplitudes interfere distructively if

$$\omega_A-\omega=-(\omega_B-\omega), \tag{3}$$

and two-photon absorption vanishes under this condition. Note that Eq. (3) is also the condition of two-photon absorption [4], i.e.,

$$\omega_A+\omega_B=2\omega. \tag{4}$$

It has been argued in the literature [5,6] that interatomic interactions like dipole-dipole interaction will lead to nonvanishing $\wp$ as in the presence of dipole-dipole interaction the states $|j\rangle$ as well as energies $E_{ji}$ become dependent on dipole-dipole interaction [5]. It is also known that dipole-dipole interaction arises from the exchange of a photon between excited and unexcited atoms [7]. This suggests an interesting possibility of the *existence of two-photon absorption in two unidentical atoms in a cavity*. In a cavity the atoms interact with a common quantized cavity field and *are effectively coupled*. We would thus examine two-photon absorption by unidentical atoms in a single-mode cavity [8]. The resulting behavior will depend on the quality of the cavity. In addition, the nonclassical aspects [8] of two-photon absorption should be quite pronounced in a high-$Q$ cavity. We note that, in the case of a low-$Q$ cavity, the cavity mode can be adiabatically eliminated and the problem essentially reduces to that of free space [5] with dipole-dipole interaction. In free space the dipole-dipole interaction arises from an infinite number of continuum of modes. In a low-$Q$ cavity the leakage to the outside world is sufficient, and this coupling to the outside world provides the continuum of modes. The dipole-dipole interaction can cause two-photon absorption depending on the spatial separation of the two atoms. In a high-quality cavity we have a single mode that interacts with two atoms, and here the quantum correlations induced by the cavity field lead to two-photon absorption. It is shown in this paper that two-photon absorption arises regardless of the spatial separation of the atoms in the cavity. Thus in what follows we concentrate on two-photon absorption by unidentical atoms in a high-$Q$ cavity [9].

The organization of this paper is as follows. In Sec. II, we present basic equations for a system of two unidentical two-level atoms interacting with the single-mode cavity field. In Sec. III, we present explicit results for two-photon absorption for a variety of the input states of the field. We identify terms in two-photon absorption which are due to the cavity. In particular, we discuss quantum correlations and the nonclassical character of two-photon absorption. The collapses and revivals in oscillations of atomic inversion are studied for the cavity field initially prepared in a state with photon number distribution with finite width. We also differentiate the results for unidentical atoms from those for identical atoms.

## II. HAMILTONIAN

We consider two unidentical atoms interacting with a single-mode cavity field with annihilation and creation operators $a$ and $a^\dagger$ [10]. The Hamiltonian of the system in a frame rotating with cavity frequency is

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$$H=\hbar\kappa_A(a^\dagger|g_A\rangle\langle e_A|+a|e_A\rangle\langle g_A|)$$
$$+\hbar\kappa_B(a^\dagger|g_B\rangle\langle e_B|+a|e_B\rangle\langle g_B|)$$
$$-\hbar\Delta_A|g_A\rangle\langle g_A|-\hbar\Delta_B|g_B\rangle\langle g_B|, \qquad (5)$$

where $\kappa_A$ and $\kappa_B$ are the coupling constants. In the model, the cavity field is detuned by $\Delta_A$ and $\Delta_B$, respectively, for atoms $A$ and $B$.

As two-level atoms are considered, an atom evolves between its ground state $|g_{A,B}\rangle$ and excited state $|e_{A,B}\rangle$, where the subscripts $A$ and $B$ refer to the atoms. Let us denote by $|g_A,g_B,n\rangle$ a state of the combined atom-field system, where both of the atoms are in their ground states and $n$ photons are present in the cavity. When similar notations are used for the other combined states of the atom-field system, the total state vector at time $t$ can be written as

$$|\Psi(t)\rangle=C_1|e_A,e_B,n-2\rangle+C_2|e_A,g_B,n-1\rangle$$
$$+C_3|g_A,e_B,n-1\rangle+C_4|g_A,g_B,n\rangle, \qquad (6)$$

where $C_i$, $i=1$, 2, 3, and 4, are the time-dependent amplitudes of states. With use of Eqs. (5) and (6), it is a simple matter to write down the equations of motion for the amplitudes:

$$\frac{\partial C_1}{\partial t}=-i\kappa_B\sqrt{n-1}C_2-i\kappa_A\sqrt{n-1}C_3,$$

$$\frac{\partial C_2}{\partial t}=-i\kappa_B\sqrt{n-1}C_1+i\Delta_B C_2-i\kappa_A\sqrt{n}C_4,$$

$$\frac{\partial C_3}{\partial t}=-i\kappa_A\sqrt{n-1}C_1+i\Delta_A C_3-i\kappa_B\sqrt{n}C_4, \qquad (7)$$

$$\frac{\partial C_4}{\partial t}=-i\kappa_A\sqrt{n}C_2-i\kappa_B\sqrt{n}C_3+i(\Delta_A+\Delta_B)C_4.$$

These four coupled differential equations can be solved analytically. In this paper, however, for a clear study of two-photon absorption, we simplify the problem by setting $\kappa_A=\kappa_B=\kappa$. Our primary interest is in the evolution of the atoms for various detunings. For identical atoms $\Delta_A=\Delta_B$, and this case has been extensively studied in the literature [11]. We keep single-photon detuning large so as to have small single-photon absorption.

## III. TWO-PHOTON ABSORPTION

We want to consider the two-photon absorption by two unidentical atoms under the condition $\Delta_A+\Delta_B=0$. Note here that single-photon detunings are nonzero. In this case, the equations of motion (7) become

$$\frac{\partial C_1}{\partial t}=-i\kappa\sqrt{n-1}C_s,$$

$$\frac{\partial C_s}{\partial t}=-2i\kappa\sqrt{n-1}C_1+i\Delta C_a-2i\kappa\sqrt{n}C_4, \qquad (8)$$

$$\frac{\partial C_a}{\partial t}=-i\Delta C_s,$$

$$\frac{\partial C_4}{\partial t}=-i\kappa\sqrt{n}C_s,$$

where $C_s=C_2+C_3$ is the amplitude of the symmetric state, $C_a=C_2-C_3$ that of the antisymmetric state, and $\Delta_A=-\Delta_B=\Delta$. In the unidentical-atom case the evolution of the system remains the four-level-atomic system, as the antisymmetric state is strongly coupled with the symmetric state.

The equations of motion (8) are easily solved. We find that when both the atoms are initially in their ground states, the probability of the atoms being in excited states at time $t$ is

$$|C_1|^2=\left(4\kappa^2\frac{\sqrt{n(n-1)}}{z_1^2}\right)^2\sin^4\left(\frac{z_1 t}{2}\right),$$
$$z_1=\sqrt{\Delta^2+2\kappa^2(2n-1)}. \qquad (9)$$

This is a simple periodic function with the Rabi frequency $2z_1$. The evolution of the population density is plotted in Fig. 1(a) when the cavity field is initially prepared with the $n=10$ Fock state. The simple sinusoidal behavior is the result of two-photon absorption by the atoms. Note the dependence of the Rabi frequency on the excitation of the Fock state. Note that effective two-photon Hamiltonians [12] will lead to a dependence of the form $n(n-1)$ square root in the argument of the sine term. Note further that if $\Delta$ is large compared to $2\kappa^2(2n-1)$, then we effectively obtain the result obtained from second-order perturbation theory. In the absence of the cavity we replace $n$ and $n-1$ by $\alpha^2$ ($\alpha$ is the $c$ number amplitude of the field), and for two-photon absorption we obtain

$$|C_1|^2=\left(\frac{4\kappa^2\alpha^2}{z_c^2}\right)^2\sin^4\left(\frac{z_c t}{2}\right), \quad z_c\equiv\sqrt{\Delta^2+4\kappa^2\alpha^2}. \qquad (10)$$

We also note the existence of trapping states if interaction time $\tau_{\rm int}$ is such that

$$\sqrt{\Delta^2+2\kappa^2(2n-1)}\,\tau_{\rm int}=2m\pi, \qquad (11)$$

where $m$ is an integer. These are analogs of the well-known [13] trapping states for single-photon absorption.

In our case of two-photon absorption, the symmetric state is coupled to the antisymmetric state, and the antisymmetric state has a chance to be populated when $\Delta\neq 0$. To consider the coupling between symmetric and antisymmetric states, we calculate the possibility of the atoms being in the antisymmetric state having assumed that the atoms are initially in the symmetric state:

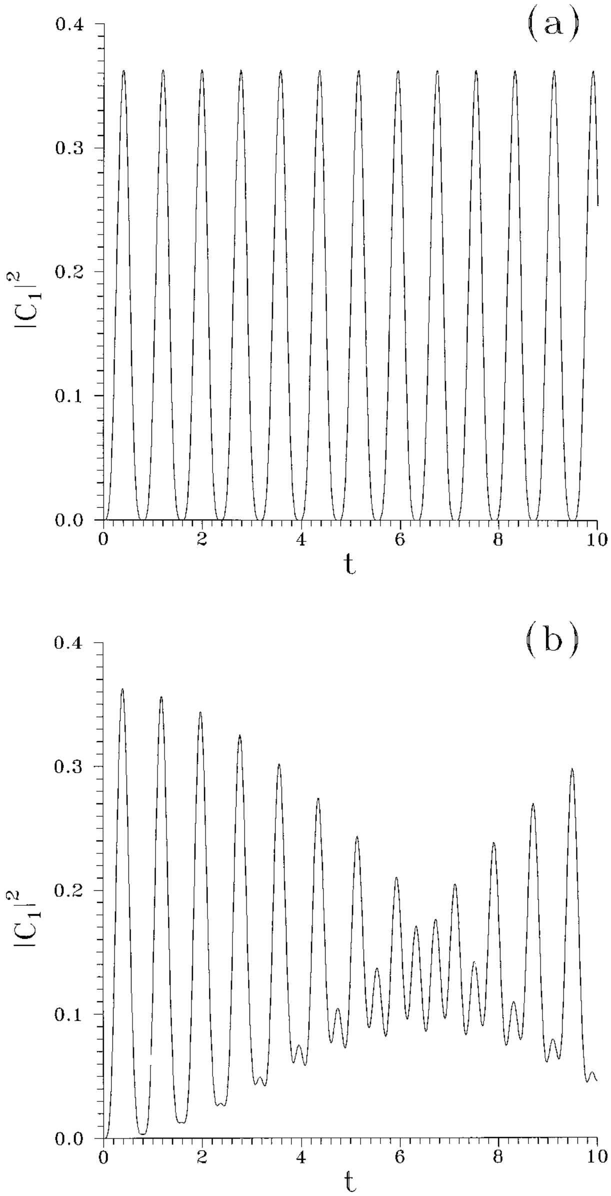


FIG. 1. Probability of both the atoms being in their excited states as a function of the interaction time $t$. The atoms were assumed to be initially in their excited states, and the cavity field initially prepared in the ten-photon Fock state. The detunings are $\Delta_A=-\Delta_B=5$ (a) and $\Delta_A=\Delta_B=5$. The $x$ axis is in units of the coupling constant $\kappa$.

$$|C_a|^2=\left(\frac{\Delta}{z_1}\right)^2\sin^2 z_1 t. \tag{12}$$

When the detuning is large, the atoms bounce back and forth between the antisymmetric and symmetric states. When the detuning is zero the antisymmetric state is decoupled, and the atomic evolution reduces to the three-level system.

We next compare the results on two-photon absorption with a system of identical atoms whose transition frequency is detuned from the cavity field, so we set $\Delta_A=\Delta_B=\Delta$. The equations of motion (7) for the two identical atoms are reduced to

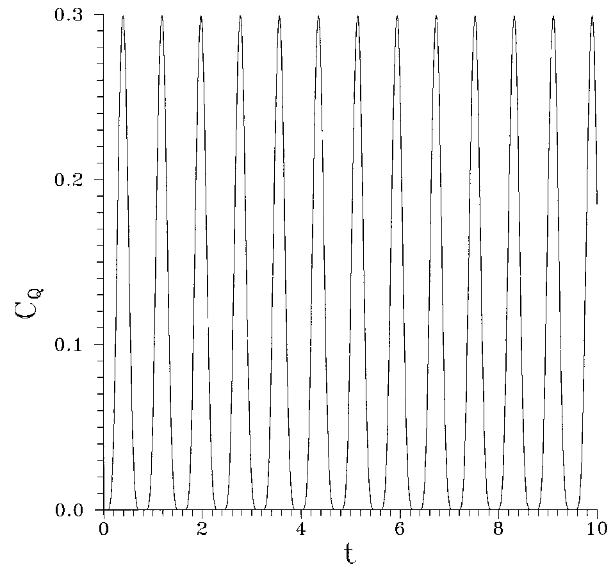


FIG. 2. Quantum correlation when the cavity field is initially prepared with the ten-photon Fock state. The detunings are $\Delta_A=-\Delta_B=5$. The $x$ axis is in units of the coupling constant $\kappa$.

$$\begin{aligned}\frac{\partial C_1}{\partial t}&=-i\kappa\sqrt{n-1}C_s,\\ \frac{\partial C_s}{\partial t}&=-2i\kappa\sqrt{n-1}C_1+i\Delta C_s-2i\kappa\sqrt{n}C_4,\\ \frac{\partial C_4}{\partial t}&=-i\kappa\sqrt{n}C_s+2i\Delta C_4.\end{aligned} \tag{13}$$

For identical atoms the two-photon resonance condition becomes identical to the one-photon resonance condition. When the atomic transition is resonant with the cavity field, the amplitude of both the atoms in the excited state is

$$C_1=-\frac{\sqrt{n(n-1)}}{2n-1}(1-\cos\Omega t), \tag{14}$$

where the resonant Rabi frequency $\Omega=\sqrt{2\kappa^2(2n-1)}$. In the identical-atom case, Eqs. (13) show that the evolution is the same as that for the three-level atom interacting with a single-mode field. When the two identical atoms are initially prepared in their ground states and the cavity field in the Fock state, the time evolution of the probability $|C_1|^2$ for both the atoms being in their excited states is plotted in Fig. 1(b).

If the detuning is chosen to be larger than the resonant Rabi frequency, i.e., $\Delta^2 \gg \Omega^2$, Eqs. (13) are approximated to have three eigenvalues, $i\Delta$, $2i\Delta$, and $-2i\kappa^2(n-1)/\Delta$. As the eigenvalue $2i\kappa^2(n-1)/\Delta\approx 0$, we notice that the three eigenvalues are nearly evenly spaced. The probability of the atoms being their excited states is

$$\begin{aligned}|C_1|^2\approx{}&2\left(\frac{\kappa}{\Delta}\right)^4 n(n-1)\\ &\times\left[3-4\cos\Delta t\,\cos\frac{\kappa^2}{\Delta}(n-1)t+\cos 2\Delta t\right].\end{aligned} \tag{15}$$

The evolution is determined by the interference between the second and third terms in the brackets. The beating occurs because there are three evenly spaced eigenvalues which are caused by the stepwise transitions from the atomic ground-ground state to the excited-excited state. In Fig. 1(b) we can see that this analogy applies even when $\Delta^2 \approx \Omega^2$. Clearly there are differences between two-photon absorption in a system of identical and unidentical atoms and these become *pronounced* for large times.

If the cavity field is not quantized, the product of each atom being in its excited state is the same as the probability of both the atoms being in their respective excited states. However, if the cavity field is quantized, the probability of the other atom to be excited is influenced by the one atom having been excited. We define the quantum correlation $C_Q$ as

$$C_Q \equiv \langle |e_A, e_B\rangle\langle e_A, e_B|\rangle - \langle |e_A\rangle\langle e_A|\rangle\langle |e_B\rangle\langle e_B|\rangle. \quad (16)$$

This value is *zero* when a *classical field* is considered to interact with the atoms. For our model, the quantum correlation is plotted in Fig. 2 when the cavity is initially prepared in the Fock state. The quantum correlations are found to be quite pronounced. In fact, Fig. 2 shows that $C_Q$ is of the same order as $|C_1|^2$.

It may also be noted that the probabilities of only a single-atom excitation are different for the cases of $\Delta_A = -\Delta_B$ and $\Delta_A = \Delta_B$. Assuming the atoms are initially in their ground states, the probability of atom $A$ being in its excited state while leaving atom $B$ in its ground state, at time $t$, is

$$|C_2|^2 = \frac{2\kappa^2 n \Delta^2}{z_1^4}\left[2\sin^2\frac{z_1}{2}t + \left(\frac{\kappa}{\Delta}\right)^2 (2n-1)\sin^2 z_1 t\right], \quad (17)$$

when $\Delta_1 = -\Delta_2 = \Delta$. If the detunings are chosen as $\Delta_A = \Delta_B$, the probability amplitude of only atom $A$ being excited is the inverse-Laplace transform of

$$\tilde{C}_2(z) = \frac{-i\kappa\sqrt{n}\, z}{z^3 - 3i\Delta z^2 + 2[\kappa^2(2n-1) - \Delta^2]z - 4i\Delta\kappa^2(n-1)}. \quad (18)$$

In the limit for large $n$, the denominator in Eq. (18) can be factored. When there is only a single photon present initially in the cavity, both evolutions (17) and (18) will show simple sinusoidal oscillations. In general, however, evolution (17) behaves differently from evolution (18). The evolutions can also be compared with the case if the second atom were absent. In this case the probability $P_e$ of the atomic excitation at time $t$ is

$$P_e = \frac{4\kappa^2 n}{\Delta^2 + 4\kappa^2 n}\sin^2\left(\frac{\sqrt{\Delta^2 + 4\kappa^2 n}\;\; t}{2}\right). \quad (19)$$

Note the distinction among the three results (17)–(19).

### Collapses and revivals

The Jaynes-Cummings model is well known for collapses and revivals in the evolution of atomic inversion when the cavity is initially in a coherent state. When the coherent field of the mean photon number $\bar{n}$ is initially in the cavity, the probability of two-photon absorption is calculated as the Poissonian-weighted sum of $|C_1|^2$:

$$P_1 = \mathrm{e}^{-\bar{n}} \sum_{n=0}^{\infty} \frac{\bar{n}^n}{n!} |C_1|^2. \quad (20)$$

When $\Delta_A = -\Delta_B$, Fig. 3(a) shows the collapse and revival character of two-photon absorption. Notice the first few full revivals. The well-defined half-revivals are also clearly seen. These half-revivals arise from the fourth power of sine term in $|C_1|^2$. When $\Delta_A = \Delta_B$, Fig. 3(b) shows that the oscillations do not revive as fully as in Fig. 3(a). The stepwise transitions between the ground-ground and excited-excited states bring about extra frequency dependence which makes the revivals less pronounced.

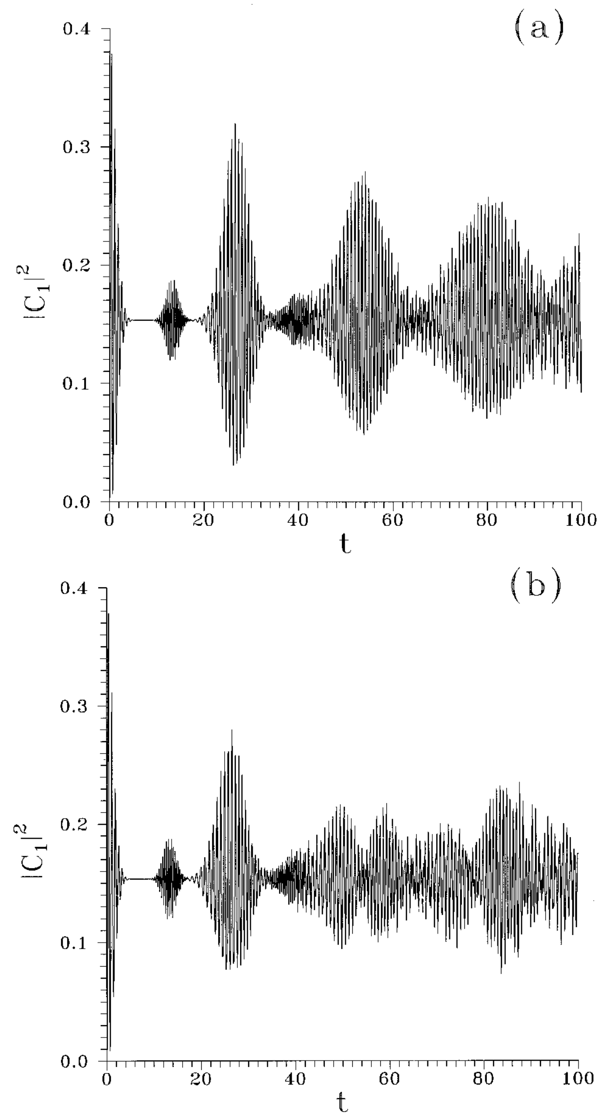


FIG. 3. Probability of both atoms being in their excited states as a function of the interaction time $t$. The atoms were assumed to be initially in their ground states, and the cavity field initially prepared in the coherent state $\bar{n} = 10$. The detunings are (a) $\Delta_A = -\Delta_B = 5$ and (b) $\Delta_A = \Delta_B = 5$. The $x$ axis is in units of the coupling constant $\kappa$.

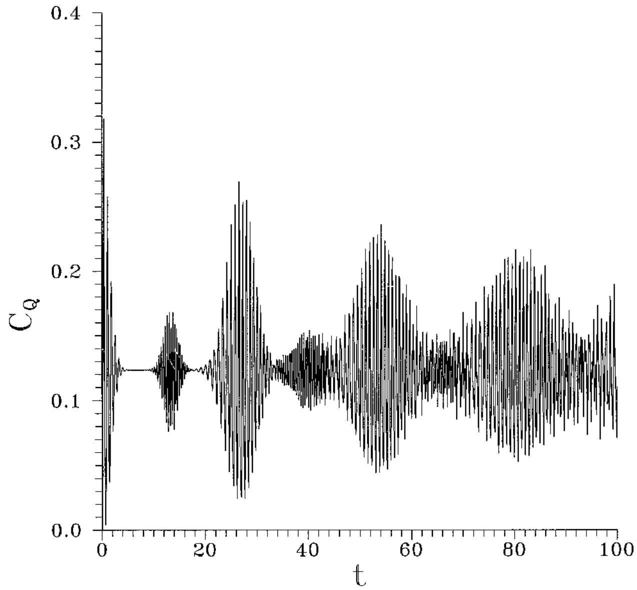


FIG. 4. Quantum correlation when the cavity field is initially prepared in a coherent state of the mean photon number $\bar{n}=10$. The detunings are $\Delta_A=-\Delta_B=5$. We can see that the quantum correlation also shows collapses and revivals. The $x$ axis is in units of the coupling constant $\kappa$.

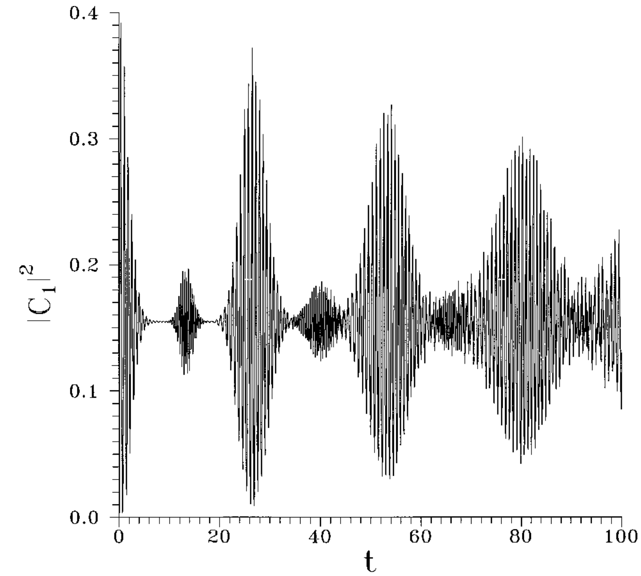


FIG. 5. Effect of sub-Poissonian statistics on the collapse and revival phenomena in two-photon excitation for $\bar{n}=10$, $\Delta_A=-\Delta_B=5$, and $\sigma^2=\bar{n}/2$. The $x$ axis is in units of the coupling constant $\kappa$.

The quantum correlation $C_Q$ is plotted in Fig. 4 when the cavity field is in the coherent state. We still note large quantum correlations, even though the field is in a coherent state. However, the average number of photons is not large enough for semiclassical limit to apply.

We also find that the revivals and fractional revivals become much more sharp (Fig. 5) if the field in the cavity has sub-Poissonian statistics; the photon number distribution of the initial cavity field is assumed to be

$$P(n)=\frac{1}{\sqrt{2\pi}\sigma}\exp\left\{-\frac{(n-\bar{n})^2}{2\sigma^2}\right\},\quad \sigma=\sqrt{\bar{n}/2}. \qquad (21)$$

## IV. CONCLUSIONS

We have studied the dynamics of two unidentical atoms interacting with a single-mode field in a lossless cavity. The atomic transitions are considered to be detuned from the cavity frequency. When the sum of the detunings vanishes, the population density of both the atoms being in their excited states is determined by simple sinusoidal evolution for an initial Fock state cavity field. We have also found that the antisymmetric state is strongly coupled with the symmetric state for the unidentical-atom interaction. In particular, when the detuning is large, the atomic system oscillates between symmetric and antisymmetric states. The collapses and revivals of oscillations in the atomic inversion persist for a longer interaction time for the case of two-photon absorption. We have found that the atomic system is strongly correlated via cavity field when the field is quantized.

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