Coherent two-photon transitions in Rydberg atoms in a cavity with finite Q

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The effects of the cavity damping on the phenomenon of collapse and revival in the population inversion of an atom in a two-photon process are investigated by using the dressed-atom approximation. Explicit results are given for a field initially in either a coherent or a two-photon coherent state. The cavity damping is expected to have more significant effects on two-photon transitions because the vacuum-field Rabi frequency for two-photon transitions is smaller than that for single-photon transitions.

A simple model[1] to study the electrodynamics of the matter–electromagnetic-field interaction is that of an atom with infinitely sharp energy levels interacting with a single undamped mode of the field. Realization of such a system is now possible in experiments on Rydberg atoms in high-Q cavities.[2–4] In these experiments, the cavity losses, however small, need to be accounted for although the finite but long lifetime of the atomic states does not play an essential part. The effects of the leakage of photons from the cavity on the dynamical and statistical properties of the system have been studied in detail in the case when the cavity mode is on resonance with the transition between the initial atomic state and another state coupled to it by dipole coupling.[5–7] The effects of the cavity damping on two-photon transitions are expected to be more significant because the vacuum-field Rabi frequency for two-photon transitions is smaller than the corresponding frequency for single-photon transitions. The transition between the two levels in this case occurs through intermediate levels with which they have common dipole coupling. The transition to the intermediate levels is, however, nonresonant or virtual. The usefulness of Rydberg atoms in the study of such coherent two-photon processes has recently been discussed by Brune *et al.*,[8] who have also proposed an experiment to build a two-photon Rydberg-atom maser.

In the case of two-photon processes in infinitely-high-Q cavities, it is known[9,10] that there are collapses and revivals of oscillations in population inversion in the presence of a coherent or chaotic field. These collapses and revivals are both compact and regular. This is in contrast with the case of single-photon transitions in which the revivals are only partial in a coherent-state field and very irregular in a chaotic field. In this paper we study the effects of cavity damping on the dynamics of two-photon processes in a cavity with high but finite Q. We adopt the dressed-atom approximation to derive analytic results for the time dependence of atomic inversion for a variety of the initial states of the field. Explicit results are given for a field initially in either a coherent or a two-photon coherent state.

We consider an atom which has a lower state $|g\rangle$ coupled to an excited state $|e\rangle$ by dipole-allowed transitions through the intermediate states $|i\rangle$ $(i=1,2,\ldots)$. The atom interacts with the electromagnetic field in a cavity tuned to the frequency $\omega=(E_e-E_g)/2\hbar$, where E_g (E_e) is the energy of the lower (excited) state. If the frequencies $(E_i-E_g)/\hbar$ and $(E_e-E_i)/\hbar$ (where E_i is the energy of an intermediate state $|i\rangle$) are sufficiently different from the field frequency ω, then it can be shown that the transitions to the intermediate levels can be considered as virtual so that the atom acts as an effective two-level system absorbing and emitting two photons of frequency ω each at a time. The effective Hamiltonian of the system is then given by

$$H=2\hbar\omega S_z+\hbar\omega a^\dagger a+\hbar g(a^{\dagger 2}S_-+S_+a^2)\,, \tag{1}$$

where a $(a^\dagger)$ is the cavity-field annihilation (creation) operator, $S_+=|e\rangle\langle g|$, $S_-=|g\rangle\langle e|$, and $S_z=\frac{1}{2}(|e\rangle\langle e|-|g\rangle\langle g|)$. In the Hamiltonian H given by Eq. (1) we have ignored the Stark shift of the two levels resulting from the virtual transitions to the intermediate states. Further, if we assume that the photons leak from the cavity at a rate 2κ, then the master equation for the density matrix ρ of the combined system of the atom and the field is given by

$$\frac{\partial\rho}{\partial t}=-\frac{i}{\hbar}[H,\rho]-\kappa(a^\dagger a\rho-2a\rho a^\dagger+a^\dagger a\rho)\ . \tag{2}$$

The parameter g is related to the two-photon matrix element and it is essentially equal to the vacuum-field Rabi frequency for two-photon transitions. With a suitable choice of the atomic transitions and the cavity size etc., one can make it relatively large. For example, Brune *et al.*[8] have found $g\sim 4000$ sec^{-1} for $40S\rightarrow 39P\rightarrow 39S$ in rubidium. The damping parameter is $\kappa\sim\omega/2Q$. Thus if we choose $\omega\sim 100$ GHz, then κ/g is $10^7/Q\sim 10^{-1}$–10^{-3} for the Q values in the range 10^8–10^{10}. Thus, even in very good cavities, κ/g for two-photon transitions is about one to two orders bigger than the corresponding values for single-photon transitions.

To solve Eq. (2) we work in the dressed-state representation, i.e., the representation consisting of the complete set of eigenstates of H which are given by

$$
\begin{aligned}
&H\,|0,g\rangle=-\hbar\omega\,|0,g\rangle\ ,\\
&H\,|1,g\rangle=0\ ,\\
&H\,|\psi_n^\pm\rangle=\lambda_n^\pm\,|\psi_n^\pm\rangle\ ,\\
&|\psi_n^\pm\rangle=\frac{1}{\sqrt{2}}[\,|n,e\rangle\pm|n+2,g\rangle]\ ,\\
&\lambda_n^\pm=\hbar[\omega(n+1)\pm g\sqrt{(n+1)(n+2)}]\ .
\end{aligned}
\tag{3}
$$

Here, $|n,g\rangle$ and $|n,e\rangle$ are the eigenstates of $a^\dagger a$ and S_z such that

$$
\begin{aligned}
&a^\dagger a\,|n,g\rangle=n\,|n,g\rangle,\quad S_z\,|n,g\rangle=-\tfrac{1}{2}\,|n,g\rangle\ ,\\
&S_z\,|n,e\rangle=\tfrac{1}{2}\,|n,e\rangle\ .
\end{aligned}
\tag{4}
$$

Next, we got to the interaction picture by defining

$$W(t)=\exp(iHt)\rho(t)\exp(-iHt)\ , \tag{5}$$

and follow Ref. 6 to obtain an equation for $W(t)$ in the secular approximation ($\kappa \ll g$). In this approximation, the equations for the diagonal matrix elements of $W(t)$ are found to be

$$
\begin{aligned}
\langle\psi_n^\epsilon\,|\,\dot W(t)\,|\,\psi_n^\epsilon\rangle=2\kappa[\,&\Gamma_{n+1}^+\langle\psi_{n+1}^\epsilon\,|\,W\,|\,\psi_{n+1}^\epsilon\rangle\\
&+\Gamma_{n+1}^-\langle\psi_{n+1}^{-\epsilon}\,|\,W\,|\,\psi_{n+1}^{-\epsilon}\rangle\\
&-(\Gamma_n^+ +\Gamma_n^-)\langle\psi_n^\epsilon\,|\,W\,|\,\psi_n^\epsilon\rangle]\\
&(\epsilon=+,-)\ ,
\end{aligned}
\tag{6}
$$

whereas the off-diagonal elements of $W(t)$ are given by

$$\langle\psi_m^\epsilon\,|\,W(t)\,|\,\psi_n^\epsilon\rangle=\exp[-\kappa t(m+n+1)]\times\langle\psi_m^\epsilon\,|\,W(0)\,|\,\psi_n^\epsilon\rangle\quad(m\neq n)\ , \tag{7}$$

$$\langle\psi_m^+\,|\,W(t)\,|\,\psi_n^-\rangle=\exp[-\kappa t(m+n+1)]\times\langle\psi_m^+\,|\,W(0)\,|\,\psi_n^-\rangle\quad(\epsilon=+,-)\ , \tag{8}$$

where Γ's are defined as

$$\Gamma_n^\pm=(\sqrt{n+2}\pm\sqrt{n}\,)^2/4\ . \tag{9}$$

For solving the equations for the diagonal elements, we work with the following equation, which is derived from Eq. (6):

$$
\begin{aligned}
\dot F_n(t)&\equiv\langle\psi_n^+\,|\,\dot W(t)\,|\,\psi_n^+\rangle+\langle\psi_n^-\,|\,\dot W(t)\,|\,\psi_n^-\rangle\\
&=2\kappa[(n+2)F_{n+1}-(n+1)F_n]\ .
\end{aligned}
\tag{10}
$$

The solution of Eq. (10) can be obtained by following the procedure of Ref. 6, and it reads as

$$F_n(t)=\exp[-2\kappa(n+1)t]\times\sum_{l=n}^{\infty}\frac{(l+1)![1-\exp(-2\kappa t)]^{l-n}}{(l-n)!(n+1)!}p_l\ , \tag{11}$$

where p_l is the photon-number distribution function. If we assume that initially the atom is in the excited state $|e\rangle$ and the field is in a coherent state $|z\rangle$, then it follows that

$$p_l=\exp(-|z|^2)|z|^{2l}/l!\ . \tag{12}$$

Note that unlike the case of one-photon process,[6] the expression in Eq. (11) for $F_n(t)$ with p_l given by Eq. (12)

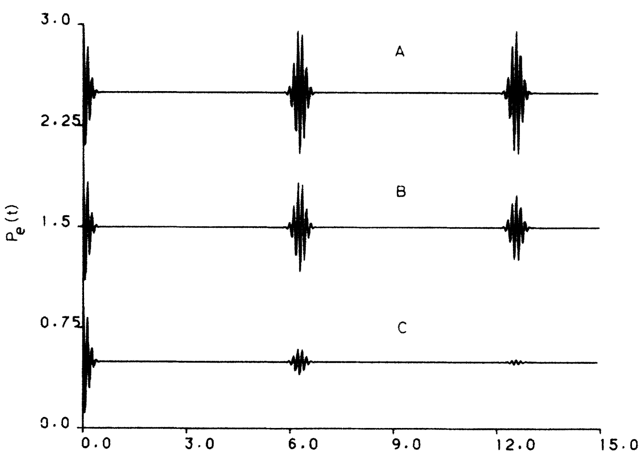

FIG. 1. Probability $P_e(t)$ of finding an atom in the excited state as a function of time for an initial coherent state with $|z|^2=50$. Curve A is for the cavity relaxation parameter $\kappa=0$ $[P_e(t)+2]$; curves B and C represent the excitation probability for $\kappa/g=0.001$ $[P_e(t)+1]$ and $\kappa/g=0.005$ $[P_e(t)]$.

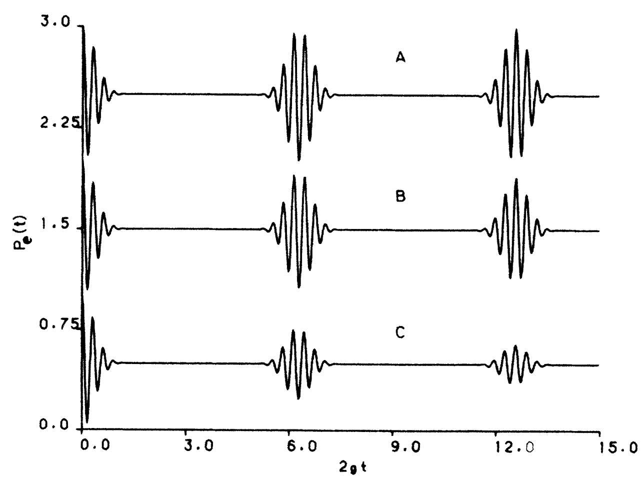

FIG. 2. Probability $P_e(t)$ of finding an atom in the excited state as a function of time for an initial squeezed state with $|z|^2=50$, $r=0.5$. Curve A is for the cavity relaxation parameter $\kappa=0$ $[P_e(t)+2]$; curves B and C represent the excitation probability for $\kappa/g=0.001$ $[P_e(t)+1]$ and $\kappa/g=0.005$ $[P_e(t)]$.

for the case of two-photon process can be summed exactly and is given by

$$F_n(t)=\frac{|z|^{2n}}{(n+1)!}\exp[-2\kappa t(n+1)]$$

$$\times\exp[-|z|^2\exp(-2\kappa t)]$$

$$\times\{n+1+|z|^2[1-\exp(-2\kappa t)]\} . \quad (13)$$

We can now evaluate the time evolution of various physical quantities. For example, the probability $P_e(t)$ of finding the atom in the excited state $|e\rangle$ may be shown to be

$$P_e(t)=\frac{1}{2}\left[\sum_{n=0}^{\infty}F_n(t)+I(t)\right] , \quad (14)$$

where

$$I(t)=\exp(-|z|^2)\sum_{n=0}^{\infty}\exp[(-2\kappa t)(n+1)]$$

$$\times\cos[2gt\sqrt{(n+1)(n+2)}]p_n . \quad (15)$$

Using Eq. (13) for $F_n(t)$, the first term in Eq. (14) reduces to

$$\sum_{n=0}^{\infty}F_n(t)=1-[1-\exp(-2\kappa t)]\exp[-|z|^2\exp(-2\kappa t)] . \quad (16)$$

The second term, $I(t)$, on the right-hand side of Eq. (14), represented by the series in Eq. (15), cannot be summed exactly. However, we can obtain an analytic expression for it by noting that in the limit $|z|^2\gg 1$, the maximum contribution to the sum in Eq. (15) comes from n near $|z|^2$ so that for $n\sim|z|^2\gg 1$ we have

$$[(n+1)(n+2]^{1/2}\simeq n+\tfrac{3}{2} , \quad (17)$$

and consequently the expression for $I(t)$ is obtained as

$$I(t)\simeq\exp(-2\kappa t)\exp\{-|z|^2[1-\exp(-2\kappa t)\cos(2gt)]\}$$

$$\times\cos[|z|^2\exp(-2\kappa t)\sin(2gt)+3gt] . \quad (18)$$

Note that $P_e(t)$ reduces to its correct limiting values of $P_e(0)=1$ and $P_e(\infty)=0$. For $\kappa=0$ we get

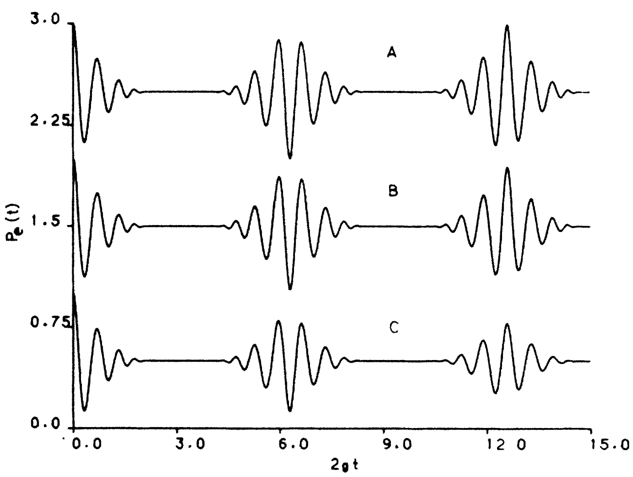

FIG. 3. Same as Fig. 2 but with $|z|^2=50$, $r=1.0$.

$$P_e(t)=1+\exp[-2|z|^2\sin^2(gt)]$$

$$\times\cos[|z|^2\sin(2gt)+3gt] . \quad (19)$$

In this case, therefore, there is a complete revival of the oscillations after their collapse. For $\kappa\neq 0$, it is seen that the oscillations damp nonexponentially. In Fig. 1 we plot $P_e(t)$ for $|z|^2=50$ and various values of κ. We have checked that for $|z|^2\gg 1$ the analytic expression derived above is in very good agreement with the results of the numerical summation of the series.

We next examine the behavior of $P_e(t)$ for the field initially in a two-photon coherent state[11] (TCS) $|\mu,z\rangle$. As is well known,[11] these states can be generated in a two-photon process and that these states are squeezed. The photon-number distribution function for a TCS is given by[11]

$$p_n=\frac{1}{2^n n!|\mu|}\frac{|\nu|^n}{|\mu|^n}H_n(z/\sqrt{2\mu\nu})H_n(z^*/\sqrt{2\mu\nu})$$

$$\times\exp\left[-|z|^2+\frac{\nu}{2\mu}(z^2+z^{*2})\right] , \quad (20)$$

where $H_n(x)$ are the Hermite polynomials and

$$\mu^2-\nu^2=1 . \quad (21)$$

Substituting Eq. (20) in Eq. (11), it can be shown that

$$\sum_{n=0}^{\infty}F_n(t)=1-E(t)[\mu^2-E^2(t)\nu^2]^{-1/2}\exp\left[\left[-|z|^2\exp(-2\kappa t)[\mu^2+\nu^2E(t)]+\frac{\mu\nu}{2}(z^2+z^{*2})[1-E^2(t)]\right]\Big/[\mu^2-\nu^2E^2(t)]\right] , \quad (22)$$

where

$$E(t)=1-\exp(-2\kappa t)\ . \tag{23}$$

Next, to evaluate $I(t)$ [Eq. (15)] by making the approximation of Eq. (17) we require p_n to be sharply peaked around the mean photon number $\bar{n}$ and by requiring that $\bar{n}\gg 1$. For p_n given by Eq. (20), we know that

$$\begin{aligned}\bar{n}&\equiv\langle a^\dagger a\rangle=|\mu z-\nu z^*|^2+\nu^2\ ,\\ (\Delta n)^2&\equiv\langle (a^\dagger a)^2\rangle-\langle a^\dagger a\rangle^2=|\mu z-\nu z^*|^2(\mu^2+\nu^2)-\mu\nu[(\mu z-\nu z^*)^2+\text{c.c.}]+2\mu^2\nu^2\ .\end{aligned} \tag{24}$$

If $|\mu z-\nu z^*|\gg\nu$ then it can be shown[12] that

$$(\Delta n)^2\sim\bar{n}[\mu^2+\nu^2-2\mu\nu\cos(2\phi)]\ , \tag{25}$$

where

$$\mu z-\nu z^*=|\mu z-\nu z^*|\exp(i\phi)\ . \tag{26}$$

The approximation of Eq. (17) can be made if $\Delta n\ll\bar{n}$, $\bar{n}\gg 1$, and in this case we find that

$$\begin{aligned}I(t)=\frac{\exp(-2\kappa t)}{\sqrt{R}}&\left\{\exp\left[-|z|^2[1-\exp(-2\kappa t)\cos(2gt+2\theta)/R]+\frac{\nu}{2\mu}(z^2+z^{*2})[1-\exp(-4\kappa t)\cos(4gt+2\theta)/R]\right]\right.\\ &\left.\times\cos\left[|z|^2\exp(-2\kappa t)\sin(2gt+2\theta)/R-\left[\frac{\nu}{2\mu}(z^2+z^{*2})\right]\exp(-4kt)\sin(4gt+2\theta)/R+3gt+\theta\right]\right\}\ ,\end{aligned} \tag{27}$$

where

$$\begin{aligned}R^2=&[\mu^2-\nu^2\exp(-4kt)\cos(4gt)]^2\\ &+\nu^4\exp(-8kt)\sin^2(4gt)\ ,\end{aligned} \tag{28}$$

$$\tan(2\theta)=\frac{\nu^2\exp(-4kt)\sin(4gt)}{\mu^2-\nu^2\exp(-4kt)\cos(4gt)}\ . \tag{29}$$

$P_e(t)$ may now be determined by substituting Eqs. (22) and (27) in Eq. (14). It may be verified that Eqs. (22) and (27) reduce to the corresponding expressions for the initial coherent state $|z\rangle$ in the limit $\mu=1$, $\nu=0$.

In Figs. 2–4 we have plotted $P_e(t)$ for an initial squeezed state having $|z|^2=50$, $\phi=0$, and for various values of squeezing parameter r [$\mu=\cosh(r)$, $\nu=\sinh(r)$] and the damping constant κ. In Fig. 2 we have $r=0.5$, and in Fig. 3 $r=1.0$. In both these cases $\Delta n\ll\bar{n}$ and the approximate analytic result of Eq. (27) is valid. Note that

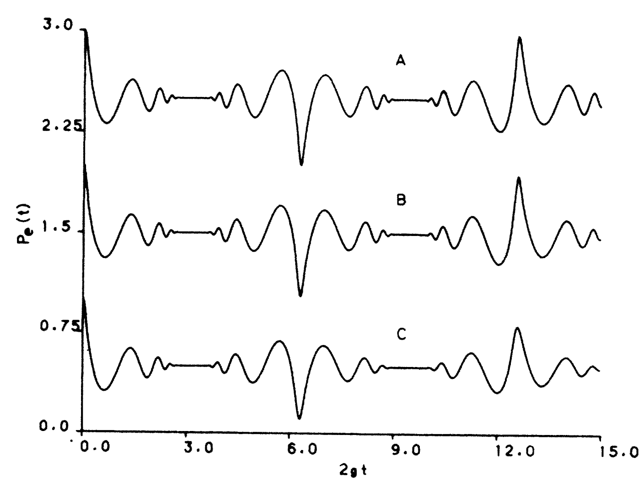

FIG. 4. Same as Fig. 2 but with $|z|^2=50$, $r=1.5$.

the time of revival t_r for $\kappa = 0$ is π/g, which is in agreement with Eq. (27). Also, as r increases there is increase in the spread of revivals. The effect of damping is also diminished with an increase in r. On the other hand, in the case of Fig. 4, where $r = 1.5$, $\Delta n \gg \bar{n}$ and in this case the analytic expression, Eq. (27), is no longer applicable. It is, however, interesting to note that the revivals in this case also are separated by the period π/g.

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