Effect of atomic motion on Rydberg atoms undergoing two-photon transitions in a lossless cavity

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The question of nonlinear transient effects similar to self-induced transparency and adiabatic following is examined for a moving Rydberg atom undergoing two-photon transition in a lossless cavity having spatial field distribution. Also, comparison is made with the case where an atom is undergoing one-photon transitions under similar conditions.

I. INTRODUCTION

Studies on Rydberg atoms have been developed so well that they allow one to prepare a two-level atom very strongly coupled to the radiation field so that the atomic coupling with a single mode becomes the dominant process in the system evolution. The exact solution of a single two-level atom interacting with a single quantized mode of the radiation field has been provided by the Jaynes-Cummings model¹ (JCM). This model has been proven to be of fundamental importance in understanding many effects in atom-radiation interaction. New effects that have been impossible to prove now fall in the range of possible detection avenues. A few of them are modification of the spontaneous emission rate of a single atom in a resonant cavity,^{2,3} the oscillatory energy exchange between an isolated atom and the cavity mode,³ and the disappearance and quantum revivals of an optical nutation signal induced on a single atom by a resonant field.⁴⁻⁶ After the advent of the superconducting cavities having a very high-quality factor O, the investigation of the interaction between a single atom and a single mode of a radiation field has become experimentally feasible.⁷ The important experiments, among others, are those that involve longer interaction times where quantum revivals have been observed.⁸ Thus the JCM is very useful in the understanding of basic matter-field interactions and also plays an important role in laser and maser theories.⁹

Recently, JCM has been studied with a different theory in which atomic motion and the effect of field structure of the cavity mode were taken into account.¹⁰ Incidentally, this provides a most elementary model for the interaction of a single atom with an electromagnetic pulse. The recent experiments^{7,8} are provided with an atomic beam passing along the axis of the rectangular or cylindrical cavity so that one can study the interaction of an atom with different cavity field modes.

The standard JCM describes an atom interacting with a constant electric field. It has been shown that when mode structure is taken into account, the nonuniform shape of the resonator mode gives rise to nonlinear transient effects in the atomic populations which are similar to self-induced transparency (SIT) and adiabatic following¹¹ (AF) respectively. These studies, however, are for an atom undergoing a one-photon transition.¹⁰ In this paper we examine these nonlinear transient effects arising from the mode structure for a moving atom undergoing two-photon transitions inside a lossless cavity.

II. THE MODEL

We assume that the atom undergoes a two-photon transition of frequency 2w between the nondegenerate states $|g\rangle$ (the ground state with energy hw_g) and $|e\rangle$ (the excited state having energy hw_e). The transitions are mediated by a single intermediate level $|i\rangle$ (energy hw_i and $hw_e > hw_i > hw_g$). The frequencies of transitions $|g\rangle \rightarrow |i\rangle$ and $|i\rangle \rightarrow |e\rangle$ are $w - \delta$ and $w + \delta$ with the coupling constants g_1 and g_2 , respectively. The Hamiltonian that describes the interaction between an effective two-level atom of states $|g\rangle$ and $|e\rangle$ and the cavity mode of frequency w can be written down as

$$H = \hbar w_{g} |g\rangle \langle g| + \hbar w_{e} |e\rangle \langle e| + \hbar wa^{\dagger}a$$
$$+ a^{\dagger}a(\beta_{2}|e\rangle \langle e| + \beta_{1}|g\rangle \langle g|)$$
$$+ g[(a^{\dagger})^{2}|g\rangle \langle e| + a^{2}|e\rangle \langle g|].$$
(1)

[The derivation of the above Hamiltonian and its validity has been discussed in considerable detail elsewhere (see Refs. 12 and 13). The main assumption behind its derivation is as follows: If the value of δ is quite large compared to the Rabi oscillation frequencies of $|g\rangle \rightarrow |i\rangle$ and $|i\rangle \rightarrow |e\rangle$, then the virtual state $|i\rangle$ can be eliminated adiabatically.] Here β_1 and β_2 are the parameters describing the intensity-dependent Stark shifts of the two levels due to the virtual transitions to the intermediate level $|i\rangle$; g is the two-photon coupling constant related to g_1, g_2 , and δ ; and $a(a^{\dagger})$ are annihilation (creation) operators for the cavity field mode. We assume further that the shape function of the cavity mode is f(z). We re-

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strict our studies for atomic motion along the z axis so that only the z dependence of the mode function would be necessary to consider. The atomic motion would be incorporated as follows:¹⁴

$$f(z) \to f(vt) , \qquad (2)$$

in which v denotes the atomic velocity. Hence, we can define our cavity mode TE_{mnp} like

$$f(vt) = \sin(p \pi vt / L) . \tag{3}$$

Here p stands for the number of half-wavelengths of the mode inside a cavity of length L. With the inclusion of the shape function of the cavity mode, the Hamiltonian of Eq. (1) can be conveniently cast in the following form:

$$H = \hbar w_{g} |g\rangle \langle g| + \hbar w_{e} |e\rangle \langle e| + \hbar wa^{\dagger}a$$
$$+ a^{\dagger}a (\beta_{2}|e\rangle \langle e| + \beta_{1}|g\rangle \langle g|)$$
$$+ [f(z)]^{2}[(a^{\dagger})^{2}|g\rangle \langle e| + a^{2}|e\rangle \langle g|].$$
(4)

The wave function of the above system can be written in the following way:

$$\psi(t) = \sum_{n} a_{n}(t) |e, n\rangle + b_{n}(t) |g, n\rangle .$$
(5)

The dynamics of the system is determined for each value of *n* by two coupled differential equations for the complex amplitudes a_n and b_{n+2} . Alternatively, we define three real quantities w_n , u_n , and v_n as follows:

$$u_{n} = 2 \operatorname{Re}(a_{n}b_{n+2}^{*}) ,$$

$$v_{n} = 2 \operatorname{Im}(a_{n}b_{n+2}^{*}) ,$$

$$w_{n} = |a_{n}|^{2} - |b_{n+2}|^{2} .$$
(6)

For simplicity we ignore Stark shifts and thus obtain a discrete set of two-photon Bloch-like equations of motion of the atom field system, due to the photon distribution in the field,

$$\frac{d}{dt}w_n = 2g\sqrt{(n+1)(n+2)}\sin^2(p\pi vt/L)v_n ,$$

$$\frac{d}{dt}v_n = \Delta u_n - 2g\sqrt{(n+1)(n+2)}\sin^2(p\pi vt/L)w_n , \quad (7)$$

$$\frac{d}{dt}u_n = -\Delta v_n .$$

Here, $\Delta = w_a - w_b - 2w$ is the detuning. We assume that the atom enters the cavity at time t=0 in the upper state $|e\rangle$ and we measure the population inversion at the transit time $t_T = L/v$ when the atom leaves the cavity again after passing p half-wavelengths of the electric field. In the literature we find mention of the density-matrix equation of the quantum theory of laser with atomic motion.^{15,16} We also find a considerable amount of discussion on two-photon Bloch equations in describing certain coherent effects such as photon echoes, optical nutation, adiabatic following,¹⁷ etc.

III. SOLUTION OF TWO-PHOTON BLOCH-LIKE EQUATIONS

A. On-resonance solution

The situation in which cavity eigenfrequency is on resonance with the atomic transition frequency is rather interesting. Here $\Delta = 0$ and we can solve exactly these equations. Now the Bloch equations reduce to two coupled equations:¹¹

$$w_n(t) = w_n(0)\cos\theta_n(t) ,$$

$$v_n(t) = -w_n(0)\sin\theta_n(t) ,$$

$$u_n(t) = 0 .$$

(8)

The function $\theta_n(t)$ can be interpreted as a tipping angle for Bloch vector $\rho_n = (u_n, v_n, w_n)$ and corresponds to the area of the square of the field amplitude the atom passes until the time t:

$$\theta_n(t) = 2g\sqrt{(n+1)(n+2)} \int_0^t d\tau [f(v\tau)]^2 .$$
 (9)

For the sinusoidal field we obtain

$$\theta_n(t) = 2g\sqrt{(n+1)(n+2)} \left[t - \frac{L}{2\pi pv} \sin(2\pi pvt/L) \right] .$$
(10)

We now examine the mode structure effect when an atom interacts with a mode with p > 1. At t=0 the atom enters the cavity at one end and follows the spatial structure of the TE_{mnp} mode and all the Bloch vectors ρ_n begin to rotate from the $\theta_n = 0$ position. The angle θ_n by which the vector $\rho_n(t)$ swings away from its initial positions $\rho_n(0)$ would always remain different for different *n*. No rephasing of Bloch vectors takes place while it completes one full wavelength of the mode structure. This is because "pulse area" [defined by Eq. (9)] does not vanish now over a wavelength of the mode. Hence we do not observe "spin or photon echo" effect, which is in contrast to the situation where an atom is undergoing one-photon transition and seeing the mode structure of the cavity.¹⁰

Another striking difference here in comparison to the situation when the atom is undergoing one-photon transition is as follows: In the experiments that measure the population inversion (W) at the exit of the cavity, one does not distinguish any difference in W due to odd or even p, which is in contrast to the one-photon transition situation, where for even p one finds SIT-type behavior¹⁰ irrespective of initial field statistics. In our case the tipping angle at time t_T when the atom leaves the cavity is given by (irrespective of whether p is odd or even)

$$\theta_n(t_T) = 2g\sqrt{(n+1)(n+2)}(L/v)$$
, (11)

a situation that is somewhat similar to the normal JCM where the electric field is considered to be uniform throughout the length of the cavity.

However, an interesting situation may arise when the atomic velocity v is such that the tipping angle $\theta_n(t_T)$ for some fixed value of n equals the integer multiple of 2π :

$$\theta_n(t_T) = 2\pi q \quad . \tag{12}$$

Now the Bloch vector ρ_n undergoes q Rabi oscillations inside the cavity and leaves it in the same position it entered. In other words, $\rho_n(t_T) = \rho_n(0)$. This behavior of the Bloch vector resembles the SIT of $2\pi q$ pulses in the theory of pulse propagation. However, the definition of pulse area is different here [see Eq. (9)]. Incidentally this behavior is analogous to the one-photon transition case.¹⁰ Suppose the input field is in the superposition of number states. Then a set of Bloch vectors ρ_n will be having all different tipping angles $\theta_n(t_T)$. For a fixed atomic velocity it is not possible to satisfy the condition (12) for all values of n simultaneously so that atomic inversion at the cavity exit is always different from the inversion at the cavity entrance. The only exception would be the number state where there is only one Bloch vector.

B. Far off-resonance condition

We can solve the above equations (7) in the adiabaticfollowing approximation under far off-resonance condi-

tions. With the analogy of Bloch vector precessing about the "torque" vector, we can explicitly write down the form of torque vector as

$$\Omega_n(t,\Delta) = \{ 2g\sqrt{(n+1)(n+2)}\sin^2(p\pi vt/L), 0, \Delta \} , \quad (13)$$

For large detunings and under the condition

$$\Delta/g \gg 2\langle n \rangle \tag{14}$$

 $(\langle n \rangle)$ is the average photon number in the field), we find that the Bloch vector ρ_n changes on the time scale Δ^{-1} as compared to the torque vector Ω_n , which changes on the time scale $L/p\pi v$. Therefore, if Ω_n changes its direction very slowly compared to ρ_n , then

$$(\Delta/g)gL/p\pi v \gg 1 . \tag{15}$$

Hence, ρ_n is precessing rapidly about Ω_n and adiabatically following this vector. We can write the solution of the equations under the adiabatic-following approximation:

$$w_n(t) = w_n(0) \frac{|\Delta|}{\{\Delta^2 + [2g\sqrt{(n+1)(n+2)}\sin^2(p\pi vt/L)]^2\}^{1/2}}$$
(16)

Under the off-resonance condition, we find behavior of the Bloch vector motion is completely different from the on-resonance condition. During the first quarter of the wavelength, all the vectors Ω_n and ρ_n change their initial value to some new value. In the second quarter of the wavelength, field strength decreases and torque vector Ω_{μ} reverse their motion and after completion of halfwavelength all arrive again simultaneously at their initial

1.4

0.8

0.2

w

values. The same motion repeatedly occurs in the subsequent half-period. So we find the rephasing of Bloch vector taking place analogous to the case of an atom undergoing a one-photon transition with mode structure included. However, rephasing does not take place in the

Thus for those experiments that measure atomic inversion behind the cavity, it is only relevant that the torque point at the cavity exit in the same direction as at the





FIG. 2. The same as Fig. 1 but curve A is for p=2.

standard JCM as it is due to the mode structure.



FIG. 3. Atomic version W at the cavity exit vs transit time t_T for a fixed detuning $\Delta = 5g$. Curve A is solution of Eq. (7) for p=1 cavity eigenmode and curve B is for standard JCM. In both cases the field is initially in the coherent state with mean $\langle n \rangle = 10$.

cavity entrance:

$$\Omega_n(t_T, \Delta) = \Omega_n(0, \Delta) . \tag{17}$$

In the limit of very large detuning and large transit times we find all Bloch vectors ρ_n remain unchanged in their direction during transit through the cavity. In other words, they exit in the same direction as at the entrance. So the atom leaves the cavity in the same state in which it entered, whether or not the cavity mode has an even or odd number p of half-wavelengths.

C. Numerical results

Exact numerical integration of Eqs. (7) is shown in Figs. 1 and 2 for p=1 (i.e., p odd) and p=2 (p even), respectively, along with normal JCM results. The quantity depicted in these plots is the inversion $(W = \sum_{n} w_{n})$ of an effective two-level atom undergoing two-photon transition after passing the cavity. We have plotted this quantity keeping its importance in the current experiments. In both figures we assume the cavity is sustaining a coherent-state field with $\langle n \rangle = 10$. We show in these figures the dependence of the inversion on the cavity detuning for a fixed atomic velocity $v = gL/\pi$. We do not find any transparency of even modes (p even) when $\Delta = 0$ in contrast to the case when the atom is undergoing onephoton transition.¹⁰ So the behavior of an atom undergoing two-photon transition at exact resonance $\Delta = 0$ is similar to the standard JCM result. However, at the offresonance condition, the mode structure leads to strong deviations from the standard JCM. As soon as detuning is large enough to satisfy condition (15), the inversion adiabatically follows the field profile inside the cavity. The normal JCM still predicts oscillatory behavior for large detuning. However, oscillations are compact as compared to the one-photon case.

In Fig. 3 we have plotted inversion with respect to the transit time $t_T = L/v$ for a fixed large detuning $\Delta = 5g$, in order to show the discrepancy for large detuning between normal JCM and JCM with mode structure. We observe here that for very fast atoms $(t_T \ll \pi/g)$ the torque Ω_n changes rapidly and mode structure effects give some oscillatory behavior. However, as soon as transit time t_T is large enough to satisfy the condition for adiabatic following, the population of the upper level is no longer affected by the passage through the cavity. The standard JCM still predicts collapses and revivals in the population inversion (which are compact and complete) because torque vector Ω_n changes steplike at the cavity entrance and exit so that the Bloch vector ρ_n can no longer follow it adiabatically.

IV. CONCLUSIONS

With the help of JCM we have studied nonlinear transient effects that are due to atomic motion and mode structure of the field for an atom undergoing two-photon transitions. The striking features of our investigations have been compared with the normal JCM model of two-photon transition. We find at far off-resonance the rephasing occurs at every node of the field—an effect which is clearly due to the mode structure of the field. A necessary condition for the rephasing due to adiabatic following is, however, that the field change continuously and at a rate that is small compared to the detuning.

Also, a very important difference between the situation where the atom is undergoing one-photon transition and the situation where it is undergoing two-photon transition has been brought out. On exact resonance it is the pulse area that determines the dynamics of the system. For the one-photon case we find the pulse area vanishes whenever the field completes one wavelength; hence we observe self-induced-transparency (SIT)-like behavior irrespective of the initial-field statistics. However, in the two-photon case the area under the square of the field amplitude never vanishes so we do not come across SIT except for some number state.

In current experiments where population inversion of the exit of the cavity is an accessible quantity, these results may be useful to discern between the type of transition undergone in the atom under the on-resonance condition.

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- ¹E. T. Jaynes and F. W. Cummings, Proc. IEEE 51, 89 (1963).
- ²E. M. Purcell, Phys. Rev. **69**, 681 (1946); D. Kleppner, Phys. Rev. Lett. **47**, 233 (1981).
- ³S. Haroche, in *Atomic Physics*, edited by D. Kleppner and F.

- ⁴A. Faist, E. Geneux, P. Meystre, and A. Quattropani, Helv. Phys. Acta **45**, 956 (1972).
- ⁵J. H. Eberly, N. B. Narozhny, and J. J. Sanchez-Mondragon,

Pipkins (Plenum, New York, 1980), Vol. 7.

Phys. Rev. Lett. 44, 1323 (1980); Y. I. Yoo and J. H. Eberly, Phys. Rep. 118, 239 (1985), and references therein.

- ⁶P. L. Knight and P. M. Radmore, Phys. Rev. A 26, 676 (1982).
- ⁷D. Meschede, H. Walther, and G. Muller, Phys. Rev. Lett. **54**, 551 (1985).
- ⁸G. Rempe, H. Walther, and N. Klein, Phys. Rev. Lett. **58**, 353 (1987).
- ⁹P. Fillipowicz, J. Javanainen, and P. Meystre, Opt. Commun. 58, 327 (1986).
- ¹⁰R. R. Schlicher, Opt. Commun. 70, 97 (1989).
- ¹¹L. Allen and J. H. Eberly, Optical Resonance and Two-Level

Atom (Wiley, New York, 1975).

- ¹²R. R. Puri and R. K. Bullough, J. Opt. Soc. Am. B 5, 2021 (1988).
- ¹³A. Joshi and R. R. Puri, Opt. Commun. 75, 189 (1990).
- ¹⁴M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., Laser Physics (Addison-Wesley, Reading, MA, 1974).
- ¹⁵D. M. Kim, M. O. Scully, and W. E. Lamb, Jr., Phys. Rev. A 2, 2534 (1970).
- ¹⁶D. D. Riska and S. Stenholm, J. Phys. A 3, 536 (1970).
- ¹⁷D. Grichkowsky, M. M. T. Loy, and P. F. Liao, Phys. Rev. A 12, 2514 (1978).