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Unitary transformation and the dynamics of a three-level atom interacting with two quantized field modes

Moorad Alexanian
Department of Physics, University of North Carolina at Wilmington, Wilmington, North Carolina 28403

Subir K. Bose
Department of Physics, University of Central Florida, Orlando, Florida 32816
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Starting from a three-level atom coupled to two modes of radiation field, we derive a Raman-coupled Hamiltonian by a unitary transformation, evaluated perturbatively in coupling constants. The Rabi oscillation frequency and the collapse and revival times of the atomic coherence are found to have strikingly different photon-intensity dependence than those found previously.

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I. INTRODUCTION

The Jaynes-Cummings model [1] of a two-level atomic system coupled to a single-mode radiation field is known to exhibit interesting optical phenomena, such as the collapse and revival of Rabi oscillations of the atomic coherence. Experiments with Rydberg atoms in microwave cavities [2] and optical systems [3] have confirmed the basic ideas of the Jaynes-Cumming model. The model continues to be extensively studied, and various extensions have been proposed [4].

In the presence of intense near-resonant single or multimode fields, nonlinear photon interactions become important. Phenomenological Hamiltonians which include two-photon couplings have been introduced by a number of authors [5]. The description of two-photon transitions in Raman-type scattering requires the introduction of an intermediate level, i.e., one considers a three-level system interacting with two modes, a pump, and a Stokes mode. Models describing Raman processes have been proposed by Gerry and Eberly [6] and Cardimona *et al* [7]. The Raman interaction Hamiltonian is obtained by using "adiabatic elimination" of the intermediate level, which produces an effective two-level system having two-photon couplings.

In this paper we show that an effective two-level Raman coupled model of the kind obtained by Gerry and Eberly and Cardimona *et al.* by adiabatic elimination can instead be obtained by a suitable unitary transformation, evaluated perturbatively in the coupling constants. This procedure also yields intensity-dependent Stark shift terms in the Hamiltonian, which are included in the calculation of the atomic population dynamics. The inclusion of the Stark effects produces a strikingly different Rabi oscillation frequency than the usual $\{n_1(n_2+1)\}^{1/2}$ behavior for the two-mode case. The collapse and revival times when the Stark effects are taken into consideration exhibit different intensity dependences than those found by Gerry and Eberly.

The paper is organized as follows. In Sec. II, we derive the two-level Raman-coupled model by unitary transformation. In Sec. III, we find the "dressed-state" eigenfunctions and eigenvalues of the full Hamiltonian. In Sec. IV, we present calculations of the population inversion and the transition probability of the atom from the ground to the excited state. The details of these calculations are given in the Appendix. In Sec. V, we discuss the effects of higher-order coupling terms in the Hamiltonian. Finally, we summarize our results and make concluding remarks in Sec. V.

II. HAMILTONIAN AND THE UNITARY TRANSFORMATION

We consider a three-level system of energies E_1, E_2, and E_3 in the so-called Λ configuration as shown in Fig. 1. The system interacts with two modes of the radiation field—a pump mode of frequency ω_1 and a Stokes mode of frequency ω_2. The Hamiltonian of the system can be written as

$$H=\sum_{i=1}^{3}E_i\sigma_{ii}+\hbar\omega_1 a_1^\dagger a_1+\hbar\omega_2 a_2^\dagger a_2+\hbar g_{12}(a_1\sigma_{21}+a_1^\dagger\sigma_{12}) +\hbar g_{23}(a_2^\dagger\sigma_{32}+a_2\sigma_{23}) , \quad (1)$$

where σ_{ij} are atomic operators given by $\sigma_{ij}=|i\rangle\langle j|$, $i,j=1,2,3$. The creation and annihilation operators of mode 1 are denoted, respectively, by $a_1^\dagger$ and a_1—the corresponding operators for mode 2 are denoted by $a_2^\dagger$ and

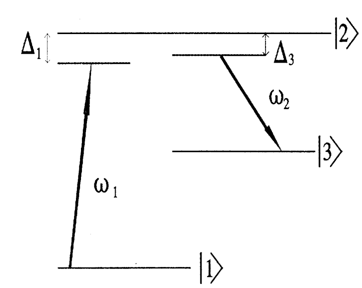

FIG. 1. Three-level atom in the Λ configuration.

a_2. Levels 1 and 2 are coupled by a dipole-coupling constant g_{12}; similarly g_{23} is the coupling constant for the $2\rightarrow 3$ transition. There is no direct coupling between levels 1 and 3. It is convenient to write the first term of Eq. (1) as

$$\sum_{i=1}^{3} E_i\sigma_{ii}=\tfrac{1}{3}(E_1+E_2+E_3)+\tfrac{1}{3}(2E_3-E_1-E_2)(\sigma_{33}-\sigma_{11}) -\tfrac{1}{3}(E_3+E_1-2E_2)(\sigma_{22}-\sigma_{11}) \, . \qquad (2)$$

We now introduce a unitary transformation

$$U=\exp(S) \, , \qquad (3)$$

where

$$S=\alpha(a_1\sigma_{21}-a_1^\dagger\sigma_{12})+\beta(a_2\sigma_{23}-a_2^\dagger\sigma_{32}) \qquad (4)$$

and α,β are constants which will be specified later.

The unitary transformation acting on the atomic and photon variables will lead to a picture of "dressed" operators, which is appropriate for the description of an atom in intense fields. The transformations cannot be found exactly; instead, we will evaluate these perturbatively and keep contributions to second order in coupling constants in the transformed Hamiltonian. Denoting the transformed operator of X by X', we have

$$X'=e^{s}Xe^{-S}=X+[S,X]+(1/2!)[S,[S,X]]+\cdots \, . \qquad (5)$$

Following this procedure we obtain the following expressions for the transformed operators:

$$a_1'=a_1+\alpha\sigma_{12}+\frac{\alpha^2}{2}a_1(\sigma_{22}-\sigma_{11})-\frac{\alpha\beta}{2}a_2\sigma_{13} \, , \qquad (6)$$

$$a_2'=a_2+\beta\sigma_{32}-\frac{\beta^2}{2}a_2(\sigma_{33}-\sigma_{22})-\frac{\alpha\beta}{2}a_1\sigma_{31} \, , \qquad (7)$$

$$\sigma_{12}'=\sigma_{12}+\alpha a_1(\sigma_{22}-\sigma_{11})-\beta a_2\sigma_{13} \, , \qquad (8)$$

$$\sigma_{23}'=\sigma_{23}-\alpha a_1^\dagger\sigma_{13}-\beta a_2^\dagger(\sigma_{33}-\sigma_{22}) \, , \qquad (9)$$

$$\begin{aligned}\sigma_{22}'-\sigma_{11}'=&(\sigma_{22}-\sigma_{11})-2\alpha a_1\sigma_{21}-2\alpha a_1^\dagger\sigma_{12}-\beta a_2\sigma_{23}\\&-\beta a_2^\dagger\sigma_{32}-(2\alpha^2+\beta^2)\sigma_{22}\\&-2\alpha^2a_1^\dagger a_1(\sigma_{22}-\sigma_{11})+\beta^2a_2^\dagger a_2(\sigma_{33}-\sigma_{22})\\&+\tfrac{3}{2}\alpha\beta(a_2^\dagger a_1\sigma_{31}+a_1^\dagger a_2\sigma_{13}) \, ,\end{aligned} \qquad (10)$$

$$\begin{aligned}\sigma_{33}'-\sigma_{11}'=&(\sigma_{33}-\sigma_{11})-\alpha a_1\sigma_{21}-\alpha a_1^\dagger\sigma_{12}+\beta a_2\sigma_{23}\\&+\beta a_2^\dagger\sigma_{32}+(\beta^2-\alpha^2)\sigma_{22}-\alpha^2a_1^\dagger a_1(\sigma_{22}-\sigma_{11})\\&-\beta^2a_2^\dagger a_2(\sigma_{33}-\sigma_{22}) \, .\end{aligned} \qquad (11)$$

The remaining transformations $a_1^{\dagger\prime}$, $a_2^{\dagger\prime}$, σ_{21}', and σ_{32}' can be obtained from the above by Hermitian conjugation. Note that σ_{12}' and σ_{23}' need to be evaluated only to first order in coupling constants. Using Eqs. (6)–(11) in Eqs. (1) and (2), we obtain the transformed Hamiltonian in the "dressed" form.

We now choose the constants α and β appropriately. If these are chosen as

$$\alpha=g_{12}/\Delta_1, \quad \beta=g_{23}/\Delta_3 \, , \qquad (12)$$

where Δ_1 and Δ_3 are the two detuning parameters defined by

$$\hbar\Delta_1=E_2-E_1-\hbar\omega_1, \quad \hbar\Delta_3=E_2-E_3-\hbar\omega_2 \, , \qquad (13)$$

we find that the terms which are linear in the field operators vanish. It is interesting that a similar result, albeit for a single-mode field, was obtained by Puri and Bullough [5] by adiabatic elimination of bilinear products of field and atom operators. With the above choice of α and β, the Hamiltonian becomes

$$\begin{aligned}H'=&\sigma_{11}E_1+\sigma_{33}E_3+\sigma_{22}\left[E_2+\hbar\frac{g_{12}^2}{\Delta_1}+\hbar\frac{g_{23}^2}{\Delta_3}\right]+\hbar\frac{g_{12}^2}{\Delta_1}a_1^\dagger a_1(\sigma_{22}-\sigma_{11})+\hbar\frac{g_{23}^2}{\Delta_3}a_2^\dagger a_2(\sigma_{22}-\sigma_{33})\\&+\hbar\omega_1a_1^\dagger a_1+\hbar\omega_2a_2^\dagger a_2-\frac{\hbar g_{12}g_{23}}{2}\left[\frac{1}{\Delta_1}+\frac{1}{\Delta_3}\right](a_1^\dagger a_2\sigma_{13}+a_2^\dagger a_1\sigma_{31}) \, .\end{aligned} \qquad (14)$$

If we assume that level 2 is very far off resonance and set the occupation number operator equal to zero, i.e., $\sigma_{22}=0$, and consider only one detuning parameter, i.e., $\Delta_1=\Delta_3=\Delta$, we obtain

$$\begin{aligned}H'=&\sigma_{11}\left[E_1-\hbar\frac{g_{12}^2}{\Delta}a_1^\dagger a_1\right]+\sigma_{33}\left[E_3-\hbar\frac{g_{23}^2}{\Delta}a_2^\dagger a_2\right]\\&+\hbar\omega_1a_1^\dagger a_1+\hbar\omega_2a_2^\dagger a_2\\&-\frac{\hbar g_{12}g_{23}}{\Delta}(a_1^\dagger a_2\sigma_{13}+a_2^\dagger a_1\sigma_{31}) \, .\end{aligned} \qquad (15)$$

In our calculations, we need not make the assumption that $\sigma_{22}=0$, since these terms do not contribute when acting on atomic states $|1\rangle$ and $|3\rangle$. We note that the intensity-dependent Stark effect terms $\sigma_{11}(\hbar g_{12}^2/\Delta)a_1^\dagger a_1$ and $\sigma_{33}(\hbar g_{23}^2/\Delta)a_2^\dagger a_2$ have emerged from our procedure. Except for a factor of 2 in the last term and the Stark effect terms, the above Hamiltonian is the same as that obtained by Gerry and Eberly with their adiabatic elimination procedure. The discrepancy of the factor of 2 will be resolved later in the text. In the subsequent discussion, the effective two-level Hamiltonian, representing a Raman-coupled model, is treated as an exact Hamiltonian.

III. DIAGONALIZATION OF THE EFFECTIVE HAMILTONIAN

The effective two-level Hamiltonian obtained in Eq. (14) is now written as

$$H_{\rm eff}=H_A+H_F+H_{AF} \ , \tag{16}$$

where H_A, H_F, and H_{AF} are, respectively, the Hamiltonians of the atom, the field, and the interaction. We write

$$H_A=\sigma_{11}E_1+\sigma_{33}E_3 \ , \tag{17}$$

$$H_F=\hbar\omega_1 a_1^\dagger a_1+\hbar\omega_2 a_2^\dagger a_2 \ , \tag{18}$$

$$H_{AF}=-\hbar\frac{g_{12}^2}{\Delta_1}a_1^\dagger a_1\sigma_{11}-\hbar\frac{g_{23}^2}{\Delta_3}a_2^\dagger a_2\sigma_{33} -\hbar g_{12}\frac{g_{23}}{2}\left[\frac{1}{\Delta_1}+\frac{1}{\Delta_3}\right](a_1^\dagger a_2\sigma_{13}+a_2^\dagger a_1\sigma_{31}) \ . \tag{19}$$

We denote the initial and final states of the uncoupled system by $|1;n_1,n_2\rangle$ and $|3;n_1-1,n_2+1\rangle$, respectively. Here $|1;n_1,n_2\rangle$ represents a state in which the atom is in the state $|1\rangle$, while the photonic state is represented by $|n_1,n_2\rangle$, n_1 and n_2 being the photon numbers in the two modes. Similarly, $|3;n_1-1,n_2+1\rangle$ represents the state in which the atom is in the state $|3\rangle$, while there is one less photon in the pump mode and one more photon in the Stokes mode. The eigenstates of the effective Hamiltonian are represented by the linear combinations

$$\begin{aligned}|\psi_{n_1n_2}^+\rangle&=-\sin\theta_{n_1,n_2}|1;n_1,n_2\rangle+\cos\theta_{n_1,n_2}|3;n_1-1,n_2+1\rangle \ ,\\ |\psi_{n_1,n_2}^-\rangle&=\cos\theta_{n_1,n_2}|1;n_1,n_2\rangle+\sin\theta_{n_1,n_2}|3;n_1-1,n_2+1\rangle \ .\end{aligned} \tag{20}$$

These can be easily inverted to give

$$\begin{aligned}|1;n_1n_2\rangle&=-\sin\theta_{n_1,n_2}|\psi_{n_1,n_2}^+\rangle+\cos\theta_{n_1,n_2}|\psi_{n_1,n_2}^-\rangle \ ,\\ |3;n_1-1,n_2+1\rangle&=\cos\theta_{n_1,n_2}|\psi_{n_1,n_2}^+\rangle+\sin\theta_{n_1,n_2}|\psi_{n_1,n_2}^-\rangle \ .\end{aligned} \tag{21}$$

The eigenvalues of $H_{\rm eff}$ are given by

$$E_{n_1,n_2}^{\pm}=E_1+\frac{\hbar(\Delta_1-\Delta_3)}{2}-\hbar\frac{g_{23}^2}{2\Delta_3}+\hbar n_1\left[\omega_1-\frac{g_{12}^2}{2\Delta_1}\right] +\hbar n_2\left[\omega_2-\frac{g_{23}^2}{2\Delta_3}\right]\pm\Omega_{n_1,n_2} \ , \tag{22}$$

where

$$\Omega_{n_1,n_2}=\frac{\hbar}{2}\left[(\Delta_1-\Delta_3)^2 +2(\Delta_1-\Delta_3)\left\{\frac{n_1g_{12}^2}{\Delta_1}-(n_2+1)\frac{g_{23}^2}{\Delta_3}\right\} +\{n_1g_{12}^2+(n_2+1)g_{23}^2\} \times\left[n_1\frac{g_{12}^2}{\Delta_1^2}+(n_2+1)\frac{g_{23}^2}{\Delta_3^2}\right]\right]^{1/2} . \tag{23}$$

We also have

$$\tan\theta_{n_1,n_2} =\frac{-\hbar(\Delta_1-\Delta_3)-\hbar\frac{g_{12}^2}{\Delta_1}n_1+\hbar\frac{g_{23}^2}{\Delta_3}(n_2+1)+2\Omega_{n_1,n_2}}{\hbar g_{12}g_{23}\left[\frac{1}{\Delta_1}+\frac{1}{\Delta_3}\right]\sqrt{n_1(n_2+1)}} \ . \tag{24}$$

Let us look at the results if we set $\Delta_1=\Delta_3=\Delta$. In this case we obtain

$$E_{n_1,n_2}^{+}=E_1+\hbar\omega_1n_1+\hbar\omega_2n_2 \ , \tag{25}$$

$$E_{n_1,n_2}^{-}=E_1+\hbar\omega_1n_1+\hbar\omega_2n_2-\hbar\frac{g_{12}^2}{\Delta}n_1-\hbar\frac{g_{23}^2}{\Delta}(n_2+1) \ , \tag{26}$$

and

$$\Omega_{n_1,n_2}=\frac{\hbar}{2\Delta}[n_1g_{12}^2+(n_2+1)g_{23}^2] \ . \tag{27}$$

Furthermore, we have

$$\sin\theta_{n_1,n_2}=\frac{r\sqrt{n_2+1}}{[n_1+r^2(n_2+1)]^{1/2}} \ , \tag{28}$$

$$\cos\theta_{n_1,n_2}=\frac{\sqrt{n_1}}{[n_1+r^2(n_2+1)]^{1/2}} \ , \tag{29}$$

where $r=g_{23}/g_{12}$. $\Omega_{n_1n_2}$ is identified as the Rabi oscillation frequency which has an altogether different intensity dependence than the $[n_1(n_2+1)]^{1/2}$ dependence in the two-mode case. We may note, however, that Knight [5], as well as Puri and Bullough [5] have also found deviations from the $n^{1/2}$ dependence of Ω in single-mode two-photon processes.

IV. DYNAMICS OF POPULATION INVERSION

We use the density matrix to investigate the dynamics of our system. Since at $t=0$ the states are "bare," we write for the density operator

$$\rho(0)=\sum_{m_1m_2n_1n_2}C_{m_1m_2n_1n_2}|1;m_1,m_2\rangle\langle 1;n_1,n_2| \ . \tag{30}$$

The time-dependent density operator of the system is calculated in a straightforward manner. The result is

$$\rho(t)=\sum_{m_1m_2n_1n_2} C_{m_1m_2n_1n_2}\exp\left[it\left\{(n_1-m_1)\left[\omega_1-\frac{g_{12}^2}{2\Delta_1}\right]+(n_2-m_2)\left[\omega_2-\frac{g_{23}^2}{2\Delta_3}\right]\right\}\right]$$
$$\times[B_{m_1m_2}(t)|1;m_1,m_2\rangle+A_{m_1m_2}(t)|3;m_1-1,m_2+1\rangle]$$
$$\times[\langle 1;n_1,n_2|B^*_{n_1n_2}(t)+\langle 3;n_1-1,n_2+1|A^*_{n_1n_2}(t)] , \quad (31)$$

where

$$A_{m_1m_2}(t)=i\sin\left[\frac{\Omega_{m_1m_2}t}{\hbar}\right]\sin(2\theta_{m_1m_2}) ,$$
$$B_{m_1m_2}(t)=\cos\left[\frac{\Omega_{m_1m_2}t}{\hbar}\right]+i\sin\left[\frac{\Omega_{m_1m_2}t}{\hbar}\right]\cos(2\theta_{m_1m_2}) . \quad (32)$$

We now calculate a general expression for $W(t)$, the population inversion, for $\Delta_1\neq\Delta_3$. $W(t)$ is given by

$$W(t)=\rho_{33}^A(t)-\rho_{11}^A(t) , \quad (33)$$

where ρ_{33}^A and ρ_{11}^A are reduced density operators of the atom, i.e.,

$$W(t)=\sum_{l_1,l_2=0}^{\infty}\langle 3;l_1,l_2|\rho(t)|3;l_1,l_2\rangle-\sum_{l_1,l_2=0}^{\infty}\langle 1;l_1,l_2|\rho(t)|1;l_1,l_2\rangle . \quad (34)$$

The evaluation of Eq. (34) is rather involved. We quote the result here and the details are provided in the Appendix. $W(t)$ is given by

$$W(t)=-1+\sum_{n_1,n_2=0}^{\infty} C_{n_1n_2n_1n_2}\frac{\left[\hbar g_{12}g_{23}\left[\frac{1}{\Delta_1}+\frac{1}{\Delta_3}\right]\sqrt{n_1(n_2+1)}\right]^2}{2\Omega_{n_1n_2}^2}\sin^2\left[\frac{\Omega_{n_1n_2}t}{\hbar}\right] . \quad (35)$$

Let us look at the result for $\Delta_1=\Delta_3=\Delta$. In this case $\Omega_{n_1n_2}$ is given by Eq. (27). We obtain

$$W(t)=-1+\sum_{n_1n_2=0}^{\infty} C_{n_1n_2n_1n_2}\frac{8g_{12}^2g_{23}^2n_1(n_2+1)}{[n_1g_{12}^2+(n_2+1)g_{23}^2]^2}\sin^2\left[\frac{n_1g_{12}^2+(n_2+1)g_{23}^2}{2\Delta}t\right] . \quad (36)$$

The polynomial factor in Eq. (36) assumes its maximum value of 2 for $n_1g_{12}^2=(n_2+1)g_{23}^2$. For field modes in coherent states, $C_{n_1n_2n_1n_2}$ is given by the product of Poissonian distributions with a maximum at $n_1=\bar{n}_1$, $n_2=\bar{n}_2$. Therefore, these maxima coincide if $\bar{n}_1g_{12}^2=(\bar{n}_2+1)g_{23}^2$.

Finally, the probability of finding the atom in the state $|3\rangle$ given that at $t=0$ it was in the state $|1\rangle$ is found to be

$$P_3(t)=\sum_{n_1n_2=0}^{\infty} C_{n_1n_2n_1n_2}\left\{\hbar g_{12}g_{23}\left[\frac{1}{\Delta_1}+\frac{1}{\Delta_3}\right]\frac{\sqrt{n_1(n_2+1)}}{2\Omega_{n_1n_2}}\right\}^2\sin^2\left[\frac{\Omega_{n_1n_2}t}{\hbar}\right] . \quad (37)$$

V. HIGHER-ORDER EFFECTS

We have carried out the evaluation of H' to third order in the coupling constants with the aid of Eq. (5). The second-order expression for H' is given by Eq. (14), and the third-order term is

$$H_3'=-\frac{\hbar g_{12}}{3}\left[4\frac{g_{12}^2}{\Delta_1^2}a_1^\dagger a_1+\frac{g_{23}^2}{\Delta_3}\left[\frac{3}{\Delta_1}+\frac{1}{\Delta_3}\right]a_2^\dagger a_2\right]a_1\sigma_{21}$$
$$-\frac{\hbar g_{23}}{3}\left[4\frac{g_{23}^2}{\Delta_3^2}a_2^\dagger a_2+\frac{g_{12}^2}{\Delta_1}\left[\frac{3}{\Delta_3}+\frac{1}{\Delta_1}\right]a_1^\dagger a_1\right]a_2\sigma_{23}$$
$$+\text{H.c.} \quad (38)$$

The constants α and β, chosen to eliminate linear field operator terms in the transformed Hamiltonian, are given by

$$\alpha=\frac{g_{12}}{\Delta_1}-\frac{g_{12}g_{23}^2}{\Delta_1^2\Delta_3}-\frac{4}{3}\frac{g_{12}^3}{\Delta_1^3}-\frac{1}{3}\frac{g_{12}}{\Delta_1}\frac{g_{23}^2}{\Delta_3^2} ,$$
$$\beta=\frac{g_{23}}{\Delta_3}-\frac{g_{23}}{\Delta_3^2}\frac{g_{12}^2}{\Delta_1}-\frac{4}{3}\frac{g_{23}^3}{\Delta_3^3}-\frac{1}{3}\frac{g_{12}^2}{\Delta_1^2}\frac{g_{23}}{\Delta_3} . \quad (39)$$

The higher-order terms in the transformed Hamiltonian give intensity-dependent coupling constants in the 1↔2 and 2↔3 transitions. The intensity-dependent coupling constants have been phenomenologically introduced earlier by Buck and Sukamar [5]. To fourth order in the coupling constants, H' contains an intensity-dependent transition between levels 1 and 3 brought about by four-photon processes.

VI. CONCLUDING REMARKS

In this paper, we have derived an effective two-level Hamiltonian having two-photon interactions, starting from a three-level system interacting with two modes of the radiation field and having one-photon interactions, using a perturbative unitary transformation. Our model describes a very general situation in which the system admits two detuning parameters Δ_1 and Δ_3. The unitary transformation also generates intensity-dependent Stark shifts. Our method of obtaining an effective two-level system is different from the adiabatic elimination procedure usually resorted to and used by Gerry and Eberly. In further contrast, we retain the dynamic Stark-shift terms in deriving the population dynamics of the atom. The Rabi oscillation frequency in our work given by Eq. (23) has a significantly different structure than that obtained previously. In the limit of $\Delta_1=\Delta_3=\Delta$, $\Omega_{n_1n_2}$ given by Eq. (27) shows a remarkably different dependence on field intensity than $[n_1(n_2+1)]^{1/2}$. This difference is entirely due to the Stark-shift terms. In the limit of $\Delta_1=\Delta_3=\Delta$ and the neglect of the Stark-shift terms, $\Omega_{n_1n_2}$ is found to be similar to that obtained by Gerry and Eberly, i.e.,

$$\Omega_{n_1n_2}=\frac{\hbar\lambda}{2}\sqrt{n_1(n_2+1)}\ ,$$

where $\lambda=2g_{12}g_{23}/\Delta$.

Let us comment on the "collapse" and "revival" times that arise from our calculations. Equation (36) shows collapse and revival as expected; however, t_R, the interval between revivals, and t_c, the collapse time, behave quite differently from those found in Ref. [6]. An analysis of collapse and revival, following the arguments of Ref. [6], gives for $\Delta_1=\Delta_3=\Delta$ and for the one-mode case $(\bar{n}_2=0)t_c^{-1}=\lambda'\bar{n}_1^{1/2}$ and $t_R=2\pi/\lambda'$, where $\lambda'=g_{12}^2/\Delta$. The above behavior of t_c and t_R is just the opposite of what was found by Gerry and Eberly, whose results are $t_c^{-1}=\lambda$ and $t_R=(2\pi/\lambda)\bar{n}_1^{1/2}$. Once again, the contrasting behavior has its origin in that we take the Stark shifts into account while Gerry and Eberly drop such terms. We also note that if g_{12}^2 and g_{23}^2 are commensurate, then Eq. (36) implies that $W(t)$ is a periodic function of t. There is total revival, viz., $W(t)=-1$, with a revival time $t_R=2\pi\Delta/a$ independent of $\bar{n}_1$ and $\bar{n}_2$, with $g_{12}^2=aI_1$ and $g_{23}^2=aI_2$, where I_1 and I_2 are positive integers with no common factors.

Although obtained by two entirely different methods, the effective interaction Hamiltonian of the Raman-coupled model, arrived at using a unitary transformation by us and by adiabatic elimination procedure by Gerry and Eberly, has the same structure in the limit of $\Delta_1=\Delta_3$, except for a factor of 2. Under the unitary transformation, both the free and the interaction parts of the Hamiltonian make contributions to the "effective interaction." The contribution from the interaction part is twice, and opposite in sign, to that coming from the free Hamiltonian making the effective interaction coupling constant $g_{12}g_{23}/\Delta$. In Gerry and Eberly's work, the "effective interaction" was obtained by adiabatic elimination using the interaction Hamiltonian alone. We have redone the calculation of Gerry and Eberly for the effective Hamiltonian by obtaining the equations of motion for the operators $a_1,a_2,\sigma_{13},\sigma_{11},\sigma_{33}$ with the aid of Eqs. (A9) and (A10) of Ref. [6]. We deduce the effective Hamiltonian which gives rise to those equations. The resulting effective Hamiltonian is in complete agreement with that obtained by us and given by Eq. (15). In fact, this is the procedure followed by Puri and Bullough in Ref. [5] for the treatment of the one-mode case where the effective coupling constant comes out to be the same as ours. Gerry and Eberly obtained their effective interaction Hamiltonian [Eq. (2.4) of Ref. [6]] by simply substituting (A9) and (A10) into their interaction Hamiltonian [Eq. (2.2b) of Ref. [6]] without regard to the equations of motion of the system. Herein lies the reason for the appearance of the extra factor of 2 in the work of Gerry and Eberly.

Gerry and Eberly assumed that the Stark-shift terms are small and hence can be neglected. Puri and Bullough [5] pointed out that these cannot be neglected if g_{12} and g_{23} are comparable since g_{12}^2/Δ, g_{23}^2/Δ, and $g_{12}g_{23}/\Delta$ then become of comparable magnitudes. Indeed in the case of ^{85}Rb Rydberg atom the $40S_{1/2}\rightarrow 39P_{3/2}$ and $39P_{3/2}\rightarrow 39S_{1/2}$ dipole matrix elements are very nearly equal and very large [8] (1500 a.u.); the value of Δ is also small, $\sim$39 MHz. Further, the Stark shifts also depend upon the photon intensities n_1 and n_2. In the experiment of Kaluzny *et al.* [2], as well as the recent experiment on the microlaser [9], average photon numbers of 10 or more were achieved. Under these circumstances, the optical Stark shifts would indeed become significant. We would like to point out that optical Stark effects are well known in the theory of effective Hamiltonians describing coherent multiphoton processes for the case of semiclassical fields interacting with a three-level system [10].

It can be shown that Gerry and Eberly's theory, upon the inclusion of the Stark-shift terms in the effective Hamiltonian, gives a Rabi frequency which is linear in the field intensities if $g_{12}=g_{23}$. To show this, let us rewrite Eq. (15) as

$$H'=\sigma_{11}E_1+\sigma_{33}E_3+\hbar\omega_1a_1^\dagger a_1+\hbar\omega_2a_2^\dagger a_2-B^\dagger B\ , \tag{40}$$

where

$$B=\left[\frac{\hbar g_{12}^2}{\Delta}\right]^{1/2}\sigma_{11}a_1+\left[\frac{\hbar g_{23}^2}{\Delta}\right]^{1/2}\sigma_{13}a_2\ . \tag{41}$$

The interaction term $B^\dagger B$ commutes with the sum of the first four terms in Eq. (40). Also $BB^\dagger$ is diagonal in field and atomic variables,

$$BB^\dagger=\frac{\hbar g_{12}^2}{\Delta}\sigma_{11}a_1a_1^\dagger+\frac{\hbar g_{23}^2}{\Delta}\sigma_{11}a_2a_2^\dagger\ . \tag{42}$$

These simplifications make the time-development operator factorize, i.e., $U(t)=U_0(t)U_I(t)$, and a straightforward calculation of the atomic inversion operator can be carried out from which the Rabi oscillation frequency can be obtained. The resulting Rabi frequency has a linear dependence on the field intensities and gives rise, in general, to quasi-periodic time behavior of the dynamical variables rather than the irregular time evolution which occurs when $\Delta_1 \neq \Delta_3$ [see Eqs. (23), (35), and (37)].

It is interesting that a similar simplification occurs in the Gerry and Eberly Hamiltonian if the Stark-shift terms are retained. Denoting the Gerry and Eberly Hamiltonian by H_{GE}, we have

$$H_{GE}=H_0-2B^\dagger B \ , \tag{43}$$

where

$$H_0=\sigma_{11}\left[E_1-\frac{\hbar g_{12}^2}{\Delta}\right]+\sigma_{33}\left[E_3-\frac{\hbar g_{23}^2}{\Delta}\right]+\hbar\omega_1 a_1^\dagger a_1 +\hbar\omega_2 a_2^\dagger a_2 \ , \tag{44}$$

and B is given by Eq. (41). Now,

$$[B^\dagger B,H_0]=\frac{\hbar^2 g_{12}g_{23}}{\Delta^2}(g_{12}^2-g_{23}^2)(a_1^\dagger a_2\sigma_{13}-a_2^\dagger a_1\sigma_{31}) \ . \tag{45}$$

Therefore, if $g_{12}^2=g_{23}^2$, then $[B^\dagger,B,H_0]=0$. The resulting Rabi frequency will be linear in photon intensities. This behavior of their Hamiltonian was not discussed by Gerry and Eberly.

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APPENDIX

We sketch here the derivation of Eq. (35) which gives the population inversion as a function of time. The reduced density operator $\rho_{33}^A(t)$ is

$$\rho_{33}^A(t)=\sum_{l_1,l_2=0}^{\infty}\langle l_1,l_2;3|\rho(t)|3;l_1,l_2\rangle =\sum_{l_1,l_2}C_{l_1+1,l_2-1,l_1+1,l_2-1}|A_{l_1+1,l_2-1}|^2 \ . \tag{A1}$$

Similarly,

$$\rho_{11}^A(t)=\sum_{l_1,l_2}\langle l_1,l_2;1|\rho(t)|l_1,l_2;1\rangle =\sum_{l_1,l_2}C_{l_1,l_2,l_1,l_2}|B_{l_1l_2}|^2 \ . \tag{A2}$$

Using Eq. (32), we get

$$W(t)=\sum_{l_1,l_2=0}^{\infty}C_{l_1+1,l_2-1,l_1+1,l_2-1}\left\{\sin^2\left[\frac{\Omega_{l_1+1,l_2-1}t}{\hbar}\right]\sin^2(2\theta_{l_1+l,l_2-1})\right\} -\sum_{l_1,l_2}C_{l_1,l_2,l_1,l_2}\left\{\cos^2\left[\frac{\Omega_{l_1,l_2}t}{\hbar}\right]+\sin^2\left[\frac{\Omega_{l_1,l_2}t}{\hbar}\right]\cos^2(2\theta_{l_1,l_2})\right\} \ . \tag{A3}$$

We rewrite the first sum in Eq. (A3) by replacing $l_1+1=n_1$, $l_2-1=n_2$. This can be cast into

$$\sum_{n_1=0}^{\infty}\sum_{n_2=0}^{\infty}C_{n_1,n_2,n_1,n_2}\sin^2\left[\frac{\Omega_{n_1,n_2}t}{\hbar}\right]\sin^2(2\theta_{n_1,n_2})-\sum_{n_1=0}^{\infty}C_{n_1,-1,n_1,-1}\sin^2\left[\frac{\Omega_{n_1,-1}t}{\hbar}\right]\sin^2(2\theta_{n_1,-1}) -\sum_{n_2=1}^{\infty}C_{0,n_2,0,n_2}\sin^2\left[\frac{\Omega_{0,n_2}t}{\hbar}\right]\sin^2(2\theta_{0,n_2}) \ . \tag{A4}$$

The two last terms in Eq. (A4) vanish in view of $\sin 2\theta_{n_1,-1}=0$ and $\sin 2\theta_{0,n}=0$ as can be seen from Eq. (24). Hence, $W(t)$ is given by

$$W(t)=\sum_{n_1,n_2=0}^{\infty}C_{n_1,n_2,n_1,n_2}\left\{\sin^2\left[\frac{\Omega_{n_1,n_2}t}{\hbar}\right]\sin^2(2\theta_{n_1,n_2})-\cos^2\left[\frac{\Omega_{n_1,n_2}t}{\hbar}\right]-\sin^2\left[\frac{\Omega_{n_1,n_2}t}{\hbar}\right]\cos^2(2\theta_{n_1,n_2})\right\} \ . \tag{A5}$$

Equation (A5) can be written as

$$W(t)=-1+2\sum_{n_1,n_2=0}^{\infty} C_{n_1,n_2,n_1,n_2}\sin^2 2\theta_{n_1,n_2} \times \sin^2\left[\frac{\Omega_{n_1,n_2}t}{\hbar}\right] . \qquad (A6)$$

From the eigenvalue equations satisfied by H_{eff}, we can derive the following result:

$$\tan^2\theta_{n_1,n_2}-1+\tan\theta_{n_1,n_2}B=0 , \qquad (A7)$$

where

$$B=\frac{(\Delta_3-\Delta_1)+\frac{g_{23}^2}{\Delta_3}(n_2+1)-\frac{g_{12}^2 n_1}{\Delta_1}}{\frac{1}{2}g_{12}g_{23}\left[\frac{1}{\Delta_1}+\frac{1}{\Delta_3}\right]\sqrt{n_1(n_2+1)}} . \qquad (A8)$$

From the identity

$$\sin^2 2\theta=\frac{4\tan^2\theta}{(\tan^2\theta-1)^2+4\tan^2\theta}$$

we obtain, using Eq. (A7),

$$\sin^2 2\theta_{n_1,n_2}=\frac{4}{B^2+4} . \qquad (A9)$$

Finally, we can write

$$\sin^2 2\theta_{n_1,n_2}=\frac{\hbar^2 g_{12}^2 g_{23}^2\left[\frac{1}{\Delta_1}+\frac{1}{\Delta_3}\right]^2 n_1(n_2+1)}{4\Omega_{n_1,n_2}^2} . \qquad (A10)$$

Substituting Eq. (A10) in Eq. (A6), we obtain Eq. (35) of the text.

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