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Dynamics of population of a four-level atom due to one- and three-photon processes

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The Schrödinger equation in the rotating-wave approximation has been solved nonperturbatively to study the temporal behavior of the occupation probabilities of different levels of a four-level atom interacting with four arbitrarily intense laser fields, such that the frequency of one of them equals the sum of the other three. The three-photon-excitation population dynamics, which is already complex, is strongly modified by one-photon excitation and detunings. Under certain conditions the three-photon excitation traps the population in the state initially populated; the population starts flowing into other states when one-photon excitation is also operative.

I. INTRODUCTION

The excitation of multilevel systems by laser fields has been a subject of much interest in recent years. Eberly and his group[1] and Shore and his collaborators[2] have studied this problem in detail. Very interesting and useful information has come out of their studies. The knowledge of the dynamics of population and its variation with system parameters is crucial to problems of present experimental interest such as for ionization, dissociation, or for selective isotope separation from the excited state. The population spectra of a three-level system driven by two laser fields, using the dressed state approach has been studied by Radmore and Knight[3] and they predicted population trapping. The effect of an additional field on the population dynamics of a three-level system excited by two laser fields has been investigated by Tsukada *et al.*[4] They found that the occupation probabilities of the three levels get strongly modified due to the interference between one- and two-photon processes.

In the present paper, we study the dynamics of population of a four-level system interacting with four laser fields of arbitary strength, where one of the laser fields which couples the ground state with the highest excited state is such that its frequency is equal to the sum of the frequencies of the other three fields. We find that in addition to the complicated population structure, some quite interesting features also appear. For certain values of the Rabi frequencies and detunings, the three-photon excitation traps the population in the initially populated state but if the one-photon excitation is also operative the population starts flowing to the other levels. In the three-level problem of Ref. 4, the additional field, on the contrary, helped the system to trap the population in the ground state which was initially populated. Here we also show that for large detuning from levels 2 and 3 (i.e., when Δ_1 and Δ_2 are large) but when one- and three-photon transitions are near or at resonance, the two-level behavior predominates as already predicted by other authors.[5]

II. MODEL HAMILTONIAN AND SCHRÖDINGER EQUATION

Figure 1 portrays a four-level atom with energies E_1,E_2,E_3,E_4 (in ascending order) interacting with four laser fields of arbitrary strengths and of frequencies ω_1, ω_2, ω_3, and ω_4. The laser frequencies ω_i, $i=1$–4, are close to those of the transition frequencies of the material system as indicated in Fig. 1. It is assumed that $\omega_4=\omega_1+\omega_2+\omega_3$. In this study we neglect collisional relaxation as well as spontaneous decay and assume that the system is driven coherently. The Hamiltonian for the system can be taken as

$$H=H_a+H_1=H_a-\vec{\mu}\cdot\vec{E}\ , \qquad (1)$$

where H_a is the unperturbed Hamiltonian for the atomic system in the absence of the radiation fields, $\vec{\mu}$ is the electric dipole moment operator, and $\vec{E}$ is the total electric field due to the laser radiations. We assume the laser fields with frequencies $\omega_1,\omega_2,\omega_3,\omega_4$ and amplitudes $\epsilon_1,\epsilon_2,\epsilon_3,\epsilon_4$, respectively, to be linearly polarized in the same direction. The total electric field can be written as

$$\vec{E}(t)=\hat{e}[\epsilon_1\cos(\omega_1 t)+\epsilon_2\cos(\omega_2 t)+\epsilon_3\cos(\omega_3 t)+\epsilon_4\cos(\omega_4 t)]\ , \qquad (2)$$

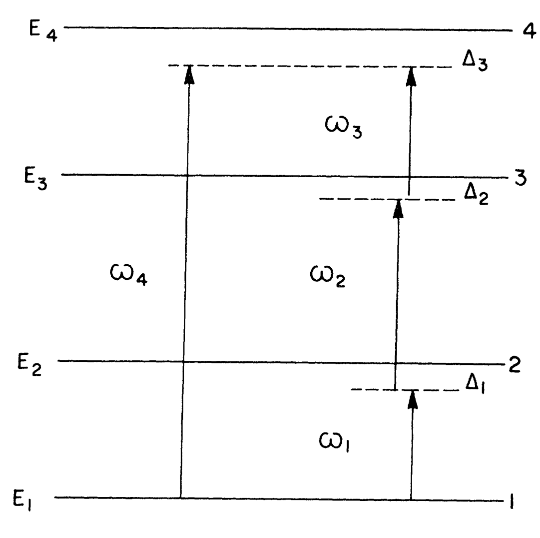


FIG. 1. Energy level diagram of a four-level system interacting with four laser fields of frequencies ω_1, ω_2, ω_3, and ω_4 as shown.

where $\hat{\epsilon}$ is the unit polarization vector. We take the initial phases of the fields to be zero and assume that these do not change with time. The wave function for the coupled atom-radiation system can be written in terms of the eigenstates of H_a as

$$|\psi(t)\rangle=\sum_{j=1}^{4} C_j(t)e^{i\Omega_j t}|j\rangle , \tag{3}$$

with

$$|1\rangle=\begin{bmatrix}1\\0\\0\\0\end{bmatrix}, \quad |2\rangle=\begin{bmatrix}0\\1\\0\\0\end{bmatrix}, \quad |3\rangle=\begin{bmatrix}0\\0\\1\\0\end{bmatrix},$$

$$|4\rangle=\begin{bmatrix}0\\0\\0\\1\end{bmatrix}, \quad \Omega_j=\frac{E_j}{h} ,$$

and $C_j(t)$ are the interaction probability amplitudes of different levels of the atom. The state function $|\Psi(t)\rangle$ can also be written as a column vector

$$|\psi(t)\rangle=\begin{bmatrix}C_1(t)e^{-i\Omega_1 t}\\C_2(t)e^{-i\Omega_2 t}\\C_3(t)e^{-i\Omega_3 t}\\C_4(t)e^{-i\Omega_4 t}\end{bmatrix} . \tag{4}$$

To study the dynamics of the population of the different levels of the atom, we need to know $C_j(t)$ which one gets from the solution of the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}|\Psi\rangle=H|\Psi\rangle . \tag{5}$$

To solve Eq. (5), we make a unitary transformation. In the transformed frame, the Hamiltonian becomes independent of time. The unitary operator which does that is

$$U(t)=e^{iAt} , \tag{6}$$

where

$$A=\begin{bmatrix}\Omega_1 & 0 & 0 & 0\\0 & \Omega_1+\omega_1 & 0 & 0\\0 & 0 & \Omega_1+\omega_1+\omega_2 & 0\\0 & 0 & 0 & \Omega_1+\omega_4\end{bmatrix} .$$

The transformed Schrödinger equation is

$$i\hbar\frac{\partial}{\partial t}|\Psi\rangle_T=H'|\Psi\rangle_T , \tag{7}$$

where

$$|\psi\rangle_T=U(t)|\Psi\rangle \tag{8}$$

and

$$H'=UHU^{-1}-i\hbar U\frac{dU^{-1}}{dt} . \tag{9}$$

The transformed Hamiltonian H' in matrix form is

$$H'=-i\begin{bmatrix}0 & -\eta_1 & 0 & -\eta_4\\-\eta_1 & \Delta_1 & -\eta_2 & 0\\0 & -\eta_2 & \Delta_2 & -\eta_3\\-\eta_4 & 0 & -\eta_3 & \Delta_3\end{bmatrix} , \tag{10}$$

where $\eta_1=|\mu_{12}\epsilon_1|/2\hbar$, $\eta_2=|\mu_{23}\epsilon_2|/2\hbar$, $\eta_3=|\mu_{34}\epsilon_3|/2\hbar$, and $\eta_4=|\mu_{14}\epsilon_4|/2\hbar$. The μ_{ij} stands for the dipole matrix element between the states $|i\rangle$ and $|j\rangle$ and the detunings are defined by the relations

$$\Delta_1=(\Omega_2-\Omega_1)-\omega_1 , \tag{11}$$

$$\Delta_2=(\Omega_3-\Omega_1)-(\omega_1+\omega_2) , \tag{12}$$

$$\Delta_3=(\Omega_4-\Omega_1)-\omega_4 . \tag{13}$$

In obtaining the elements of the matrix of Eq. (10), the rotating-wave approximation (RWA) has been used. In matrix notation, the transformed Schrödinger equation, Eq. (7), takes the form

$$\frac{d}{dt}\begin{bmatrix}C_1(t)\\C_2(t)e^{-i\Delta_1 t}\\C_3(t)e^{-i\Delta_2 t}\\C_4(t)e^{-i\Delta_3 t}\end{bmatrix}=-i\begin{bmatrix}0 & -\eta_1 & 0 & -\eta_4\\-\eta_1 & \Delta_1 & -\eta_2 & 0\\0 & -\eta_2 & \Delta_2 & -\eta_3\\-\eta_4 & 0 & -\eta_3 & \Delta_3\end{bmatrix}\begin{bmatrix}C_1(t)\\C_2(t)e^{-i\Delta_1 t}\\C_3(t)e^{-i\Delta_2 t}\\C_4(t)e^{-i\Delta_3 t}\end{bmatrix} . \tag{14}$$

To solve the above set of four coupled, first-order differential equations, we use the Laplace-transform method which converts them into a set of algebraic equations. The details of the mathematics are given in Appendix A. Our interest is in determining the occupation probabilities of each level which is defined by the relation

$$P_n(t)=|C_n(t)|^2 . \tag{15}$$

III. TIME EVOLUTION OF THE OCCUPATION PROBABILITIES OF DIFFERENT LEVELS

The mathematical details of the derivation of the expressions for the occupation probability amplitudes of different levels are presented in Appendix A. The occupation probabilities $P_n(t)=|C_n(t)|^2$ can be obtained from expressions given in Eqs. (A18), (A19), (A20), and (A21),

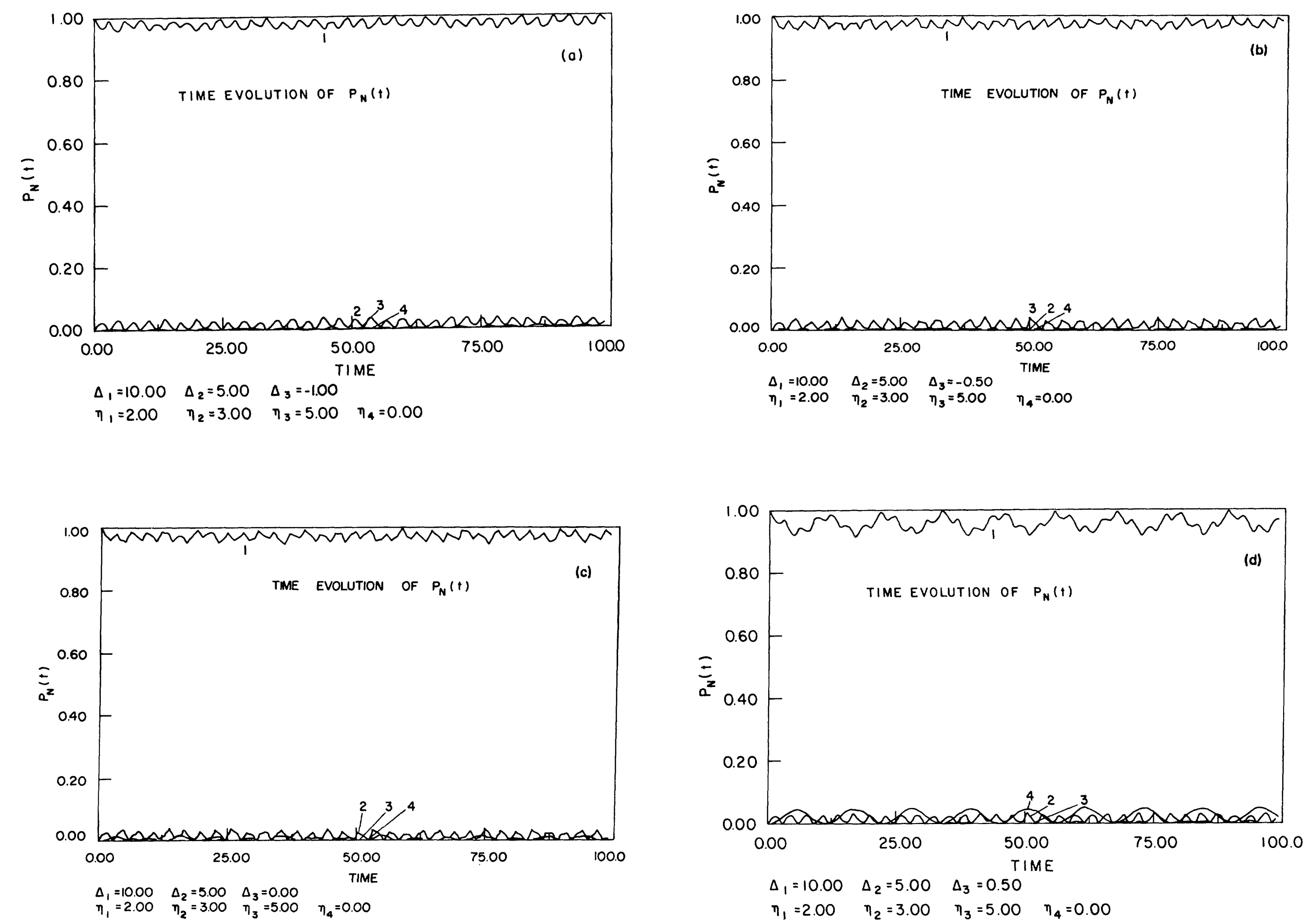


FIG. 2. Population $P_n(t)$ in the nth level as a function of time t, showing the effect of detuning Δ_3, with three-photon excitation. The calculations are carried out for the values of the parameters indicated below each figure.

for levels 1, 2, 3, and 4, respectively. The evaluation of the occupation probabilities requires the determination of the roots of the fourth-degree polynomial $F(p)=0$ [Eq. (A8)]. These roots are determined numerically. The $P_n(t)$ as a function of time has also been evaluated numerically by a computer.

For a two-level system our analysis gives the following expressions:

$$P_1(t)=1-\frac{4\eta_1^2}{\Delta_1^2+4\eta_1^2}\sin^2[(\Delta_1^2+4\eta_1^2)^{1/2}t]$$

and

$$P_2(t)=\frac{4\eta_1^2}{\Delta_1^2+4\eta_1^2}\sin^2[(\Delta_1^2+4\eta_1^2)^{1/2}t]$$

for the occupation probabilities of levels 1 and 2, respectively, and these are well-known results in the literature. For $\Delta_1=0$, there is a complete transfer of population from level 1 to level 2, whenever $2\eta_1 t=(n+\frac{1}{2})\pi$, but for any finite value of Δ_1, positive or negative, there is a partial transfer of population to the excited state. The greater the detuning, the smaller will be the probability of transfer of population to level 2.

The expressions for the occupation probabilities amplitudes for a certain level in a multilevel system interacting with more than one radiation field, as in the present case, are, in general, involved. For example, in our case the probability amplitude of occupation of level 4 is given by the expression

$$C_4(t)=\frac{e^{i\Delta_3 t}}{Q}[X_1 g_4(p_1)e^{p_1 t}+X_2 g_4(p_2)e^{p_2 t}+X_3 g_4(p_3)e^{p_3 t}+X_4 g_4(p_4)e^{p_4 t}] .$$

The symbols Q, X_j, $g_4(p_j)$ and p_j have their significance as given in Appendix A. The p_j's are all distinct imaginary quantities.

The occupation probability of fourth level is $P_4(t)=|C_4(t)|^2$ which because of the very form of $C_4(t)$ is quite involved and has interference terms. The interference effects which are sensitive to detunings and Rabi fre-

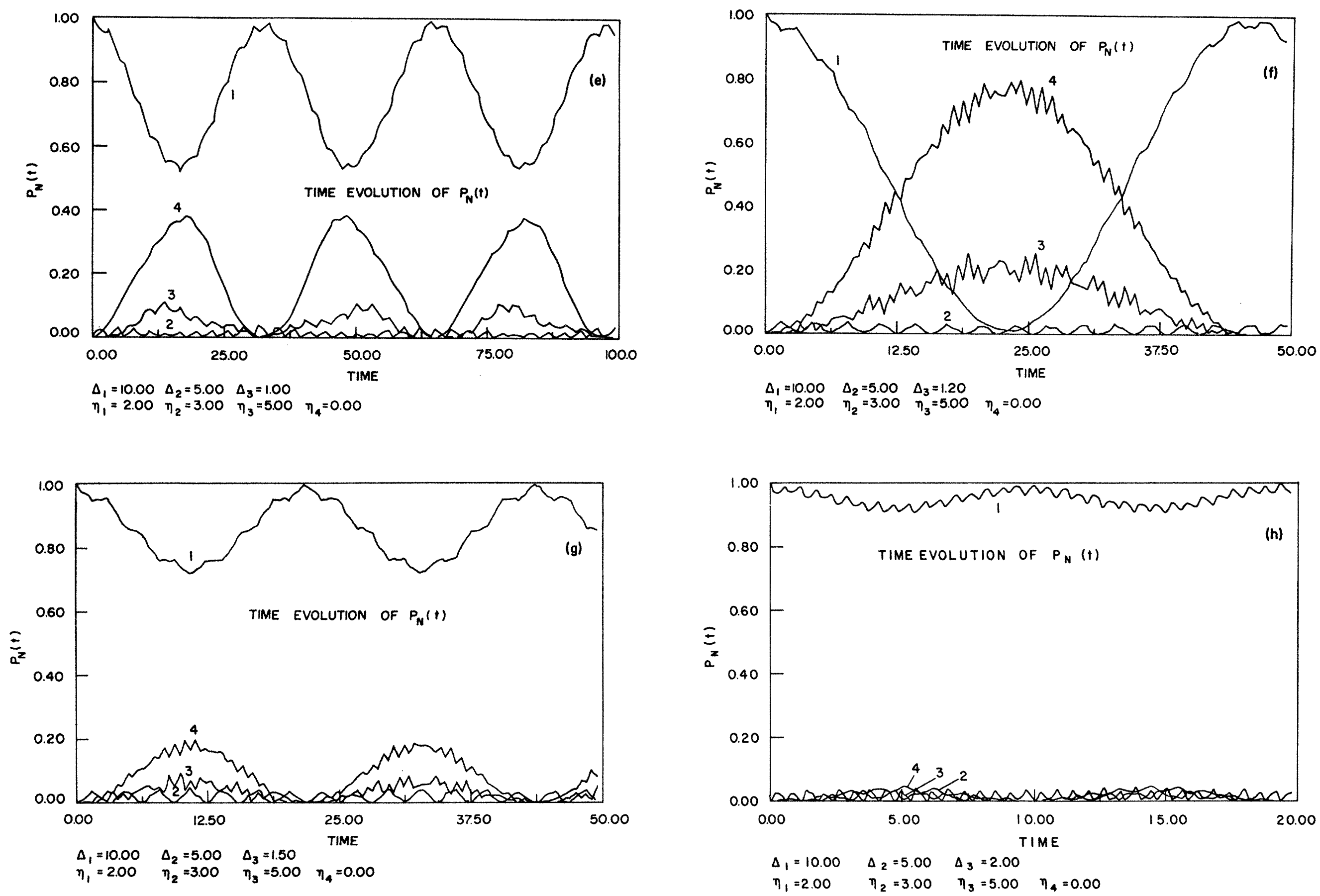


FIG. 2. (*Continued*).

quencies are reflected in the population spectra as a function of time.

Now we discuss the population dynamics of different levels with the initial condition that only the ground state is populated and all the other states are unoccupied. First we consider the case when there is no one-photon coupling between levels 1 and 4, i.e., the laser field which couples levels 1 and 4 is absent.

In Figs. 2(a)—2(h), the occupation probabilities $P_n(t)$ for the four levels are plotted as a function of time for the Rabi frequencies $\eta_1=2.0$, $\eta_2=3.0$, $\eta_3=5.0$ and detunings $\Delta_1=10$ and $\Delta_2=5$. The detunings Δ_1 and Δ_2 have been chosen to be large purposely so as to avoid populating levels 2 and 3. The value of the detuning Δ_3 is varied so as to see the effect of Δ_3 on the population dynamics of level 4. It is clear from Figs. 2(a)—2(c) that variation of Δ_3 has not produced any tangible effect on the population of level 4. But from now onward as Δ_3 is increased more and more population flows to level 4. For $\Delta_3=1.2$, we find a dramatic effect, i.e., on the average more than 40% of the population flows to level 4. But increasing Δ_3 further produces the expected result, i.e., the larger the value of Δ_3, the smaller the value of $P_4(t)$. When $\Delta_3=2$, almost all of the population stays trapped in the ground state. For the parameter chosen, the population of level 4 has shown a maxima as a function of Δ_3.

In the set of Figs. 3(a)—3(g), the value of the Rabi frequencies η_1, η_2, and η_3 have been kept the same as in the above case. Now the direct one-photon coupling between levels 1 and 4 is also operative. The Rabi frequency η_4 corresponding to the fourth field is taken to be equal to 0.30. Here again the effect of the variation of Δ_3 on the population spectra is studied. In its gross behavior, the presence of the fourth field has helped us to put more population in level 4 than its absence. For example, by comparing Fig. 2(e) with Fig. 3(e), where all other parameters are the same except η_4, we find a drastic increase in the population of level 4 when η_4 is changed from zero to 0.30.

Next we analyze the case when all the laser fields are in resonance with the atomic transition, i.e., when $\Delta_1=\Delta_2=\Delta_3=0$ and $\eta_1=2.0, \eta_2=3.0, \eta_3=5.0$. In Fig. 4(a), we set $\eta_4=0$ and in Fig. 4(b), η_4 is taken to be equal to 0.5. In both the figures we find that there is a flow of population in all the levels involved. The presence of the fourth field changes the location, height, and the number of peaks in the population spectra.

In Figs. 5(a) and 5(b), the values of the Rabi frequencies have been kept the same as in Figs. 4(a) and 4(b) but detunings have been given the values, i.e., $\Delta_1=1.0$, $\Delta_2=1.5$, and $\Delta_3=2.5$. We plot the population in each level as a function of time. As is obvious from the figures, the pop-

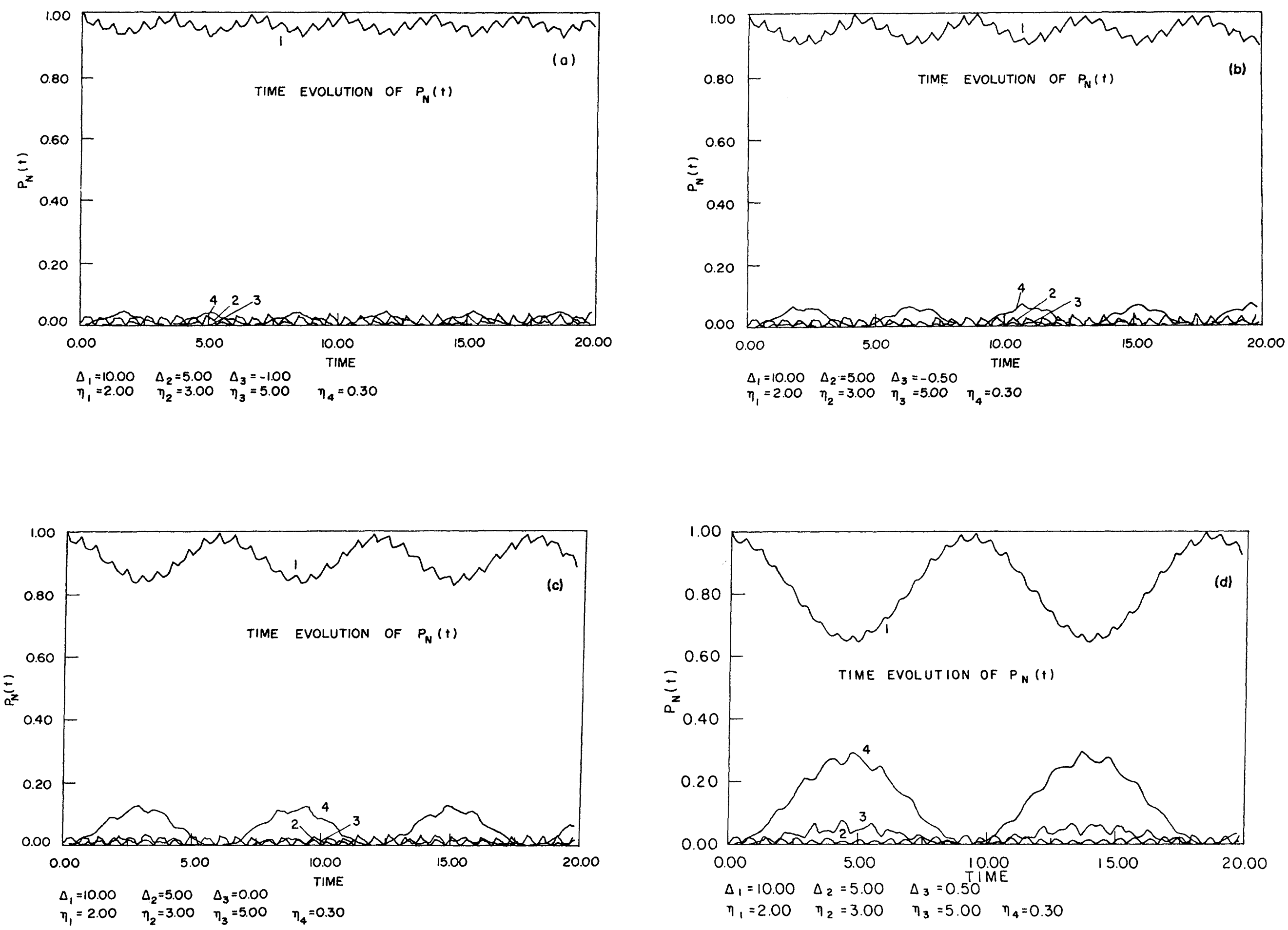


FIG. 3. Population $P_n(t)$ in the nth level as a function of time t, showing the effect of detuning Δ_3, on population spectra in the presence of both three-photon and one-photon processes.

ulation spectra is quite complex and the population is distributed in all levels.

In Fig. 6(a), we take large values of detunings, i.e., $\Delta_1=\Delta_2=10$ but $\Delta_3=0$ and Rabi frequencies $\eta_1=2.0$, $\eta_2=3.0$, $\eta_3=5.0$, and $\eta_4=0.5$. For this case we observe a two-level behavior between levels 1 and 4, as already predicted in the literature.[5] In Fig. 6(b), we set $\Delta_3=1$, while keeping the values of the other parameters the same as in Fig. 6(a). A comparison between the two figures shows that detunings Δ_3 favor flow of population to level 4 instead of hindering it.

IV. SUMMARY

In this paper we have studied numerically the population dynamics of a four-level system interacting with four electromagnetic fields of arbitrary strength. It is shown that when there is more than one laser field interacting with a multilevel system, there are interference effects which are reflected in the population spectra of different levels. The detunings have very important effects on the population spectra.

We have shown that for certain values of the parameters (Rabi frequencies and detunings) the three-photon excitation process does not succeed in taking the material system out of the ground state to the highest excited state (in this case level 4). Almost all of the population stays trapped in the ground state and the system behaves as if nothing is happening to it in spite of the presence of three laser fields interacting with the system.

In the present problem (four-level system) for the set of parameters used, the additional field (which couples level 1 to level 4) helps the population flow to level 4, while in the case of a three-level problem (Ref. 4) for their parameters the additional field hindered the flow of population to level 3.

Our study reveals that for the effective multiphoton excitation of molecules a right choice of parameters is very crucial, otherwise the system will never come out of its initial state of occupancy. So just bombarding of molecules with laser field is not sufficient to have the desired excitation.

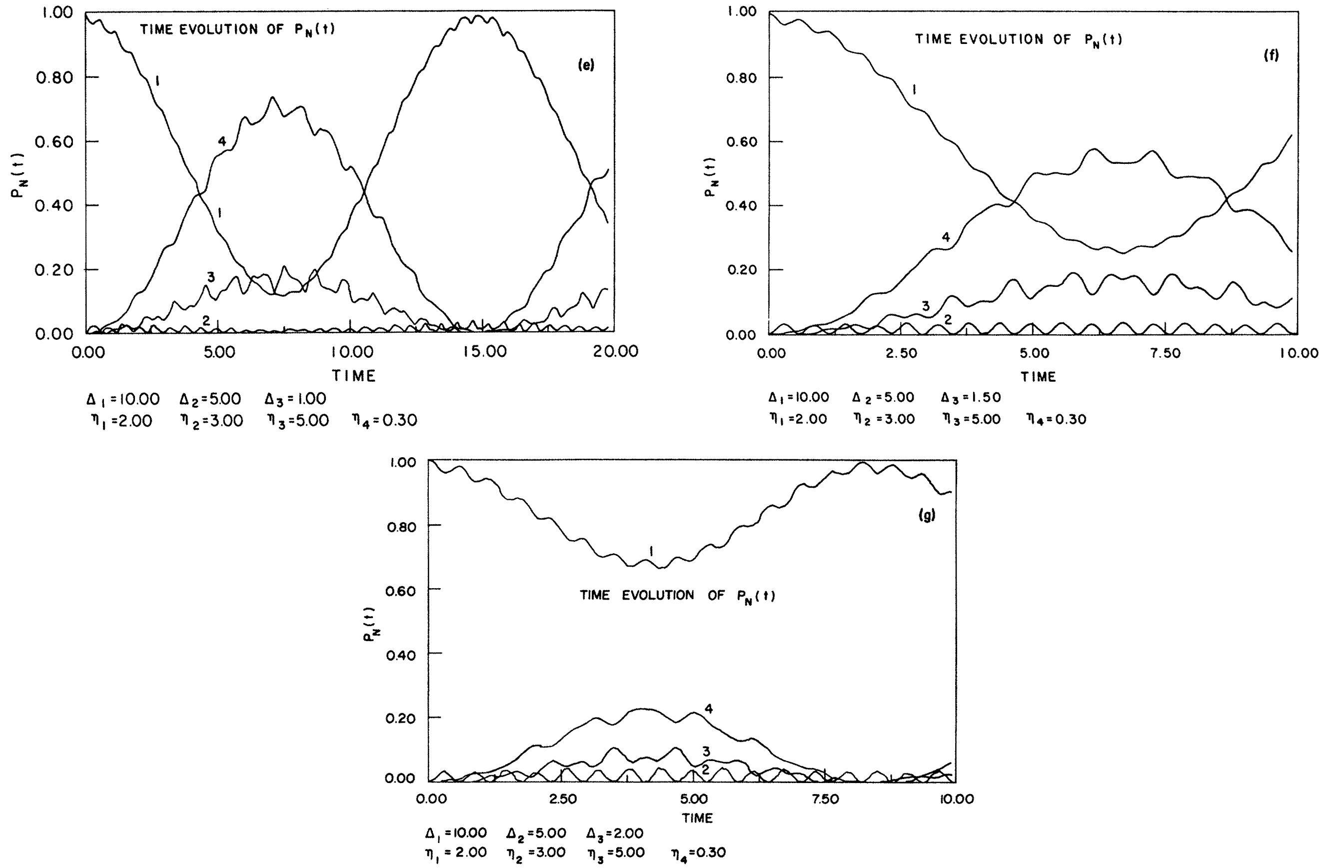


FIG. 3. (*Continued*).

APPENDIX A: CALCULATION OF PROBABILITY AMPLITUDES

In this appendix we solve the set of equations [(Eq. (14)] which have been derived in the RWA. This set of coupled first-order differential equations is solved by using the Laplace transform method. Let $C_1(p)$, $C_2(p+i\Delta_1)$, $C_3(p+i\Delta_2)$, and $C_4(p+i\Delta_3)$ be the Laplace transform of $C_1(t)$, $C_2(t)e^{-i\Delta_1 t}$, $C_3(t)e^{-i\Delta_2 t}$, and $C_4(t)e^{-i\Delta_3 t}$, respectively. Using the property of the Laplace transform of the derivatives, we get the following set of algebraic equations after the transformation:

$$pC_1(p)-C_1(0)=i\eta_1C_2(p+i\Delta_1)+i\eta_4C_4(p+i\Delta_3)\ , \tag{A1}$$

$$pC_2(p+i\Delta_1)-C_2(0)=i\eta_1C_1(p)-i\Delta_1C_2(p+i\Delta_1)+i\eta_2C_3(p+i\Delta_2)\ , \tag{A2}$$

$$pC_3(p+i\Delta_2)-C_3(0)=i\eta_2C_2(p+i\Delta_1)-i\Delta_2C_3(p+i\Delta_2)+i\eta_3C_4(p+i\Delta_3)\ , \tag{A3}$$

$$pC_4(p+i\Delta_3)-C_4(0)=i\eta_4C_1(p)+i\eta_3C_3(p+i\Delta_2)-i\Delta_3C_4(p+i\Delta_3)\ . \tag{A4}$$

The above equations after rearrangement can be put in the form of a matrix as

$$\begin{bmatrix} p & -i\eta_1 & 0 & -i\eta_4 \\ -i\eta_1 & p+i\Delta_1 & -i\eta_2 & 0 \\ 0 & -i\eta_2 & p+i\Delta_2 & -i\eta_3 \\ -i\eta_4 & 0 & -i\eta_3 & +p+i\Delta_3 \end{bmatrix} \begin{bmatrix} C_1(p) \\ C_2(p+i\Delta_1) \\ C_3(p+i\Delta_2) \\ C_4(p+i\Delta_3) \end{bmatrix} = \begin{bmatrix} C_1(0) \\ C_2(0) \\ C_3(0) \\ C_4(0) \end{bmatrix}, \tag{A5}$$

where $C_j(0)$ are the values of the probability amplitudes at the start of the interaction, i.e., when $t=0$.

We use Cramer's rule to solve the above set of equations and obtain

$$C_1(p)=\frac{f_1(p)}{F(p)}\ , \tag{A6}$$

where

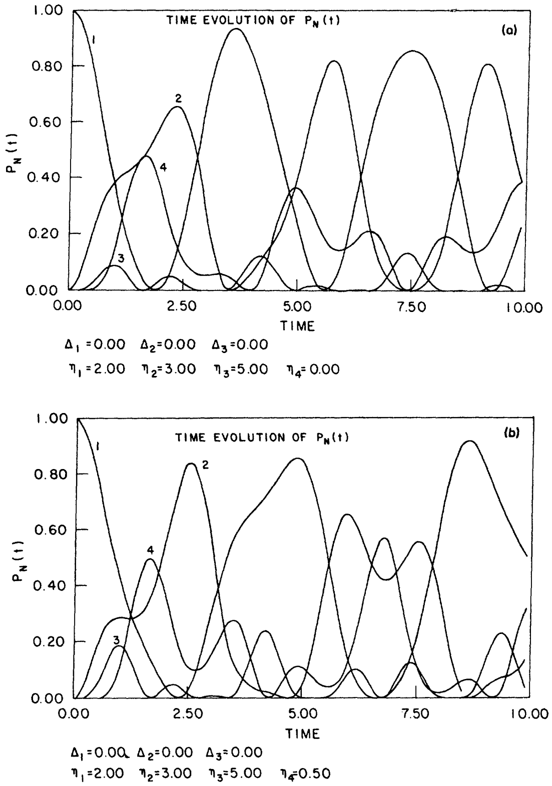


FIG. 4. Population spectra of the four-levels for the case of exact resonance with (a) $\eta_4=0$ and (b) 0.5, respectively.

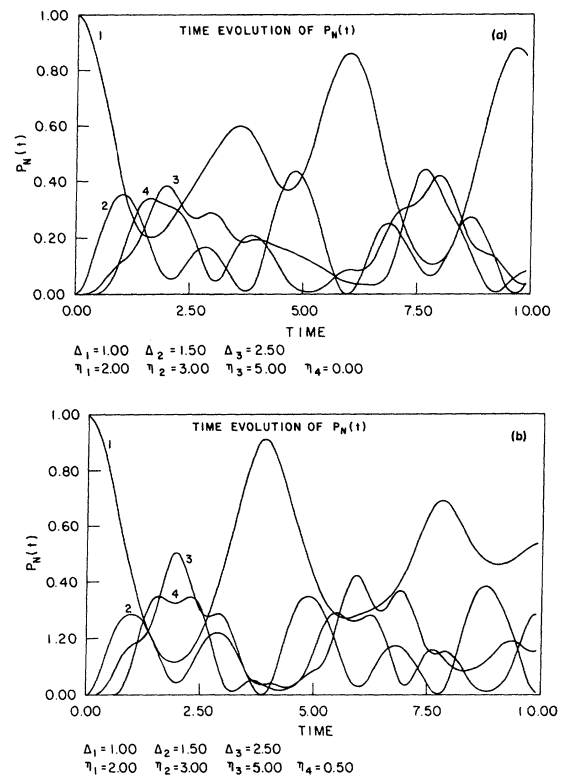


FIG. 5. Effect of η_4 on the population spectra for smaller values of Δ_1 and Δ_2.

$$\begin{aligned} f_1(p)=&C_1(0)p^3+ip^2[C_1(0)(\Delta_1+\Delta_2+\Delta_3)+C_2(0)\eta_1+C_4(0)\eta_4] \\ &+p\{C_1(0)\,|\,[\eta_2^2+\eta_3^2-(\Delta_1\Delta_2+\Delta_2\Delta_3+\Delta_3\Delta_1)]-C_2(0)(\Delta_3+\Delta_4)\eta_1-C_3(0)(\eta_1\eta_2+\eta_3\eta_4)-C_4(0)(\Delta_1+\Delta_2)\eta_4\} \\ &+i[C_1(0)(\Delta_1\eta_3^2+\Delta_3\eta_2^2-\Delta_1\Delta_2\Delta_2)+C_2(0)(\eta_3^2\eta_1-\eta_2\eta_3\eta_4-\Delta_2\Delta_3\eta_1) \\ &\quad -C_3(0)(\Delta_3\eta_1\eta_2+\Delta_1\eta_3\eta_4)+C_4(0)(\eta_2^2\eta_4-\eta_1\eta_2\eta_3-\Delta_1\Delta_2\eta_4)] \end{aligned} \tag{A7}$$

and $F(p)$ is the determinant

$$\begin{vmatrix} p & -i\eta_1 & 0 & -i\eta_4 \\ -i\eta_1 & p+i\Delta_1 & -i\eta_2 & 0 \\ 0 & -i\eta_2 & p+i\Delta_2 & -i\eta_3 \\ -i\eta_4 & 0 & -i\eta_3 & p+i\Delta_3 \end{vmatrix}$$

given by

$$\begin{aligned} F(p)=&p^4+ip^3(\Delta_1+\Delta_2+\Delta_3)+p^2[\eta_1^2+\eta_2^2+\eta_3^2-(\Delta_1\Delta_2+\Delta_2\Delta_3+\Delta_3\Delta_1)] \\ &+ip[(\Delta_2+\Delta_3)\eta_1^2+(\Delta_1+\Delta_2)\eta_4^2+\Delta_1\eta_3^2+\Delta_3\eta_2^2-\Delta_1\Delta_2\Delta_3]+[(\eta_1\eta_3)^2+(\eta_2\eta_4)^2-\Delta_1\Delta_2\eta_4^2-\Delta_2\Delta_3\eta_1^2-\eta_1\eta_2\eta_3\eta_4]\,. \end{aligned} \tag{A8}$$

The expressions for $C_2(p+i\Delta_1)$, $C_3(p+i\Delta_2)$, and $C_4(p+i\Delta_3)$ are

$$C_2(p+i\Delta_1)=\frac{f_2(p)}{F(p)}\,, \tag{A9}$$

where

$$\begin{aligned} f_2(p)=&C_2(0)p^3+ip^2[C_1(0)\eta_1+C_2(0)(\Delta_2+\Delta_3)+C_3(0)\eta_2] \\ &-p[C_1(0)(\Delta_1+\Delta_3)\eta_1-C_2(0)(\eta_3^2+\eta_4^2-\Delta_2\Delta_3)+C_3(0)\Delta_3\eta_2+C_4(0)(\eta_2\eta_3+\eta_1\eta_4)] \\ &-i\{C_1(0)[(\Delta_2\Delta_3-\eta_3^2)\eta_1+\eta_2\eta_3\eta_4]-C_2(0)\Delta_2\eta_4^2-C_3(0)(\eta_4^2\eta_2-\eta_1\eta_3\eta_4)+C_4(0)\eta_1\eta_2\Delta_2\}\,, \end{aligned} \tag{A10}$$

$$C_3(p+i\Delta_2)=\frac{f_3(p)}{F(p)}\ , \tag{A11}$$

where

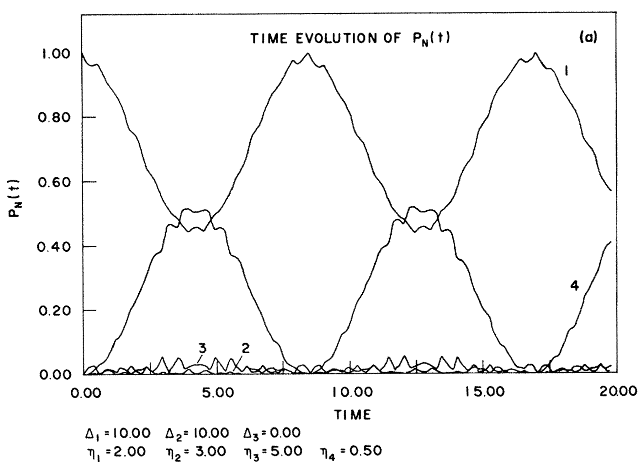
$$f_3(p)=C_3(0)p^3+ip^2[C_2(0)\eta_2+C_3(0)(\Delta_1+\Delta_3)+C_4(0)\eta_3]$$
$$+p[C_1(0)(\eta_1\eta_2+\eta_3\eta_4)-C_2(0)\Delta_3\eta_2+C_3(0)(\eta_1^2+\eta_2^2-\Delta_1\Delta_3)-C_4(0)\Delta_1\eta_3]$$
$$+i[-C_1(0)(\Delta_3\eta_1\eta_2+\Delta_1\eta_3\eta_4)+C_2(0)(\eta_2\eta_4^2-\eta_1\eta_2\eta_4)+C_3(0)(\Delta_3\eta_1^2+\Delta_1\eta_4^2)+C_4(0)(\eta_1^2\eta_3-\eta_1\eta_2\eta_4)]\ , \tag{A12}$$

and

$$C_4(p+i\Delta_3)=\frac{f_4(p)}{F(p)}\ , \tag{A13}$$

where

$$f_4(p)=C_4(0)p^3+ip^2[C_1(0)\eta_4+C_3(0)\eta_3+C_4(0)(\Delta_1+\Delta_2)]$$
$$+p[-C_1(0)(\Delta_1+\Delta_2)\eta_4-C_2(0)(\eta_2\eta_3+\eta_1\eta_4)-C_3(0)\Delta_1\eta_3+C_4(0)(\eta_1^2+\eta_2^2-\Delta_1\Delta_2)]$$
$$+i[C_1(0)(\eta_2^2\eta_4-\eta_1\eta_2\eta_3-\Delta_1\Delta_2\eta_4)-C_2(0)\Delta_2\eta_1\eta_4+C_3(0)(\eta_1^2\eta_3-\eta_1\eta_2\eta_4)+C_4(0)\eta_1^2\Delta_2]\ . \tag{A14}$$



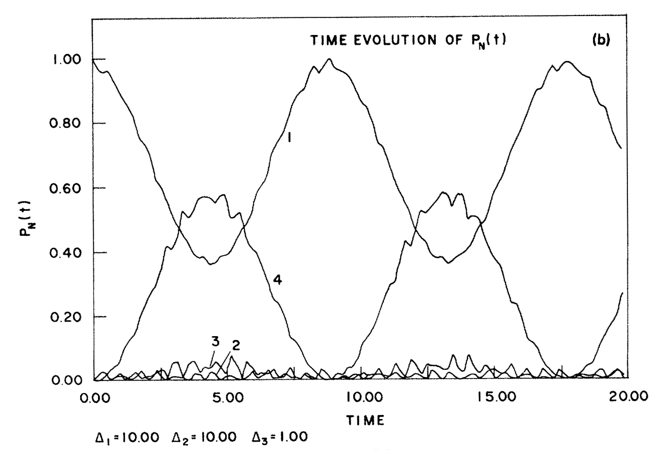


FIG. 6. Population spectra, showing the effect of detuning, when all the four laser fields are operative and Δ_1 and Δ_2 are quite large.

To obtain the time evolution of the probability amplitudes we still have to calculate the inverse Laplace transforms of Eqs. (A6), (A9), (A11), and (A13). The expressions for the probability amplitudes have the general form $f_j(p)/F(p)$, where $f_j(p)$ is a third degree polynomial in p and $F(p)$ is a fourth degree polynomial in p. $F(p)$ can be written as a product of four terms as

$$F(p)=(p-p_1)(p-p_2)(p-p_3)(p-p_4)\ , \tag{A15}$$

where p_1,p_2,p_3,p_4 are the roots of the equation $F(p)=0$. Doing the partial fraction of the expression $f_1(p)/F(p)$ and then taking the inverse Laplace transform, the expression for $C_1(t)$ reads as

$$C_1(t)=\frac{1}{Q}[X_1f_1(p_1)e^{p_1t}+X_2f_1(p_2)e^{p_2t}+X_3f_1(p_3)e^{p_3t}+X_4f_1(p_4)e^{p_4t}]\ , \tag{A16}$$

where

$$X_1=(p_2-p_3)(p_3-p_4)(p_4-p_2)\ ,$$
$$X_2=(p_1-p_3)(p_3-p_4)(p_1-p_4)\ ,$$
$$X_3=(p_1-p_2)(p_1-p_4)(p_4-p_2)\ , \tag{A17}$$
$$X_4=(p_1-p_2)(p_2-p_3)(p_1-p_3)\ ,$$
$$Q=(p_1-p_2)(p_2-p_3)(p_3-p_4)(p_1-p_3)\times(p_1-p_4)(p_4-p_2)\ ,$$

and $f_1(p)$ is given by Eq. (A7).

Replacing $f_1(p)$ by $f_2(p)$, $f_3(p)$, and $f_4(p)$ [the expressions which are given in Eqs. (A7), (A10), (A12), and (A14), respectively] in Eq. (A16), we get the expressions for $C_2(t)e^{-i\Delta_1t}$, $C_3(t)e^{-i\Delta_2t}$, and $C_3(t)e^{-i\Delta_3t}$, respectively. Here we give the expressions for the probability amplitudes $C_j(t)$, $j=1$–4 for the case when the system evolves from the ground state, i.e., $C_1(0)=1$ and $C_j(0)=0$ for $j=2,3,4$,

$$C_1(t)=\frac{1}{Q}\sum_{j=1}^{4} X_j g_1(p_j)e^{p_j t}\ , \tag{A18}$$

where

$$g_1(p)=p^3+ip^2(\Delta_1+\Delta_2+\Delta_3)$$
$$+p[\eta_2^2+\eta_3^2-(\Delta_1\Delta_2+\Delta_2\Delta_3+\Delta_3\Delta_1)]$$
$$+i(\Delta_1\eta_3^2+\Delta_3\eta_2^2-\Delta_1\Delta_2\Delta_3)\ , \tag{A19}$$

$$C_2(t)=\frac{e^{i\Delta_1 t}}{Q}\sum_{j=1}^{4} X_j g_2(p_j)e^{p_j t}\ ,$$

where

$$g_2(p)=i[p^2\eta_1+ip(\Delta_2+\Delta_3)\eta_1 -(\eta_1\eta_3^2-\eta_2\eta_3\eta_4+\Delta_2\Delta_3\eta_1)]\ ,$$

$$C_3(t)=\frac{e^{i\Delta_2 t}}{Q}\sum_{j=1}^{4} X_j g_3(p_j)e^{p_j t}\ , \tag{A20}$$

$$g_3(p)=+[(\eta_1\eta_2+\eta_3\eta_4)+i(\eta_1\eta_2\Delta_3-\eta_3\eta_4\Delta_1)]\ ,$$

and

$$C_4(t)=\frac{e^{i\Delta_3 t}}{Q}\sum_{j=1}^{4} X_j g_4(p_j)e^{p_j t}\ , \tag{A21}$$

where

$$g_4(p)=i[p^2\eta_4+ip(\Delta_1+\Delta_2)\eta_4 +(\eta_2^2\eta_4-\eta_1\eta_2\eta_3-\Delta_1\Delta_2\eta_4)]\ .$$

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