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On the population loss dynamics of a strongly excited four-level system

L. KANCHEVA, E. POPOV

Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee Boulevard, 1784 Sofia, Bulgaria

and G. GEORGIEV

Institute for Nuclear Research and Nuclear Energy, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria

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Abstract. The population dynamics of a strongly excited four-level quantum system with allowed loss out of the system from the highest state are analysed. This work has been stimulated by the paper of Cardimona *et al.* and represents a more general treatment of the problem studied by them. In particular, we show that the reduction in the ground-state population decay rate for large losses is not a consequence of the two-level behaviour but is a more general phenomenon. On the basis of numerical and analytical solutions we demonstrate that for an arbitrary excitation regime, at sufficiently large decay rate, the population loss dynamics of the four-level system is reduced to that of a slowly damped three-level system. We demonstrate also that the detailed knowledge of the coherent population dynamics enables us to predict the effect of decay rate on the population's time evolution for arbitrary choice of dynamic parameters.

1. Introduction

Details of the dynamics of multimode-laser excited N-level atoms and molecules have been reported in numerous articles and monographs [1] and, at present, considerable knowledge has accumulated. Since in strong resonance interaction of light with matter the conventional perturbation theory is no longer adequate, non-perturbative approaches are needed for interaction description which often give results far from those expected intuitively. Thus in a paper by Cardimona *et al.* (CSG) [2] a specific feature of the population dynamics of a strongly driven four-level system with allowed decay out of the system at a rate γ from the highest level has been demonstrated. CSG observed a substantial decrease in the population decay rate with increasing γ, contrary to the intuitively expected by them fast exponential decay. CSG have examined the damping of population probabilities of a ladder-like four-level atom, excited in a regime of an effective two-level atom between the ground and the uppermost level [3–5]. This enabled them to analyse the effect of the decay width γ on the system's population loss dynamics in terms of a two-level damped atom [6]. Within the two-level approximation the derivation [2] has yielded that for values of γ smaller than the effective Rabi frequency the population decays at rate γ, and, when γ is greater than Ω_{eff}, the population decay rate is $\Omega_{\text{eff}}^2/\gamma$. Because of the very small magnitude of Ω_{eff} (for the parameter values chosen by CSG, Ω_{eff} is 0·075) a drastic decrease in the ground-state population decay rate has been observed in [2].

Although at first glance this is unexpected, in fact this result is well known in the theory of resonant multiphoton ionization of atoms. It has been shown [7] that, when stimulated pumping processes dominate the dynamics (strong-field limit), the total bound-state population decays at a rate γ and, when the ionization rate is much larger than the laser intensities (weak-field limit), the population decays at a rate Ω^2/γ.

As one can see, the same decay rate constants for two extreme regimes of excitation were obtained by CSG using a two-state system model. The work of CSG [2, 6] (see also Vol. I, section 3.10 of [1]) once again demonstrates how, based on the simplest two-level model, fundamental relations and features of the light–matter interaction can be obtained. Indeed, the simplicity of the model allows greater physical insight into the ongoing processes than is generally obtained from more complicated calculations. The addition of a loss mechanism to the simple two-level system, as well as to the effective two-level system, leads to a decrease in the coherent coupling between the ground and the excited state. When the loss rate appreciably exceeds the Rabi frequency, the system is not yet in the 'Rabi regime', the coupling of states becomes insignificant and the population which initially occupied the ground state almost does not leave this state. Precisely this situation is demonstrated in figures 3 (*c*) and (*d*) in [2]. For the sets of parameters chosen there, γ exceeds Ω_{eff} by an order or more. That is why almost the entire population remains trapped on the ground level from which there is a very slow decay. This substantial reduction in the population decay rate CSG explained with an effect originating only from the two-level behaviour, ignoring any kind of multiphoton interference effects. Such an assertion is not quite correct, because it is the competition between stimulated multiphoton processes and the irreversible loss process which actually determines the temporal behaviour of population probabilities.

In the present paper we shall show that the reduction in the population decay rate is not a peculiarity of two-level behaviour but is of a more general nature. We shall demonstrate that in the general case for sufficiently large γ (exceeding the values of all dynamic parameters substantially) the uppermost level is practically decoupled from the rest of the levels. As a result, the dynamics of the four-level system are reduced to those of a weakly damped three-level system with induced losses from the third level at a rate Ω_3^2/γ.

The second question which we shall discuss is: what is a high or a low loss rate in the case of a strongly driven four-level system? If for a two-level atom a quantitative criterion for the two competitive processes can be defined, for a multilevel system with more than two coupled levels, there is no longer a simple connection between the magnitudes of the applied Rabi frequencies and the loss rate. We shall show that the character of the damping process depends not so much upon the magnitudes of the basic dynamic parameters (Rabi frequencies, resonance detunings and loss rate) but upon the relationships between them. It is precisely these relationships that make the original four-level system behave as an effective two-, three- our four-level system. That is why, in order to predict the effect of losses on the population damping dynamics, it is compulsory to take into account multiphoton processes and their interference.

In the next sections we shall present examples illustrating the population loss dynamics of a four-level system excited in different excitation regimes and shall point out some of the regularities.

2. Atom-field model and equations of motion

The model of a four-level atom as well as the notation are the same as in the paper of CSG [2], namely a ladder-like four-level atom is irradiated by three sufficiently intense near-resonant laser fields, so that spontaneous decay and other relaxation processes can be ignored. Decay out of the system only from the highest state at rate γ is allowed (figure 1).

Under these assumptions, the probability amplitudes $C_i(t)$ obey the rotating-wave approximation time-dependent Schrödinger equations with phenomenologically included decay [8]:

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \end{bmatrix} = \begin{bmatrix} 0 & \Omega_1 & 0 & 0 \\ \Omega_1 & D_1 & \Omega_2 & 0 \\ 0 & \Omega_2 & D_2 & \Omega_3 \\ 0 & 0 & \Omega_3 & D_3-\mathrm{i}\gamma \end{bmatrix}\begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \end{bmatrix}. \tag{1}$$

We would like to point out that using the notation in [2] we define the on-resonance Rabi frequencies as $\Omega_{ij}=2|\mu_{ij}E_i/h|$ and the irreversible loss as 2γ, which differs by a factor of two from the commonly used notation [8].

Our interest will be concentrated on the time evolution of the system's population $P_i(t)=|C_i(t)|^2$, being initially only on the ground state $P_1(t=0)=1$.

3. Population dynamics

3.1. *Numerical solutions*

In this section we present graphs of numerical solutions of equation (1). In the figures, together with level population histories we plot the time evolution of losses, which for concretization will be referred to as ionization $I(t)=1-\sum_i P_i(t)$.

Let us start with some simplest cases of on-resonance excitation. It is known [9, 10] that, at exact resonance conditions for all fields, the four-level atom behaves as an effective two-level atom if one of the applied Rabi frequencies is much greater than the two others. When one of the two external fields is much stronger Ω_1 (or Ω_3)$\gg\Omega_2$, Ω_3 (or Ω_1), the system's population is localized only between the first and the second levels and, when the intermediate field is much more intense

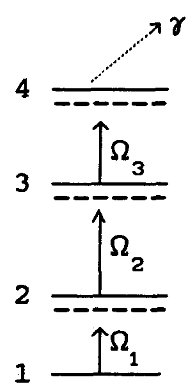
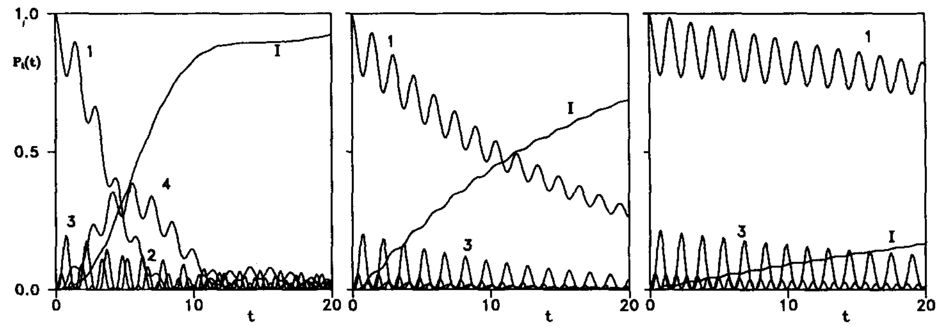


Figure 1. Ladder-like four-level atom pumped by three laser fields and allowed losses out of the system from the highest level.

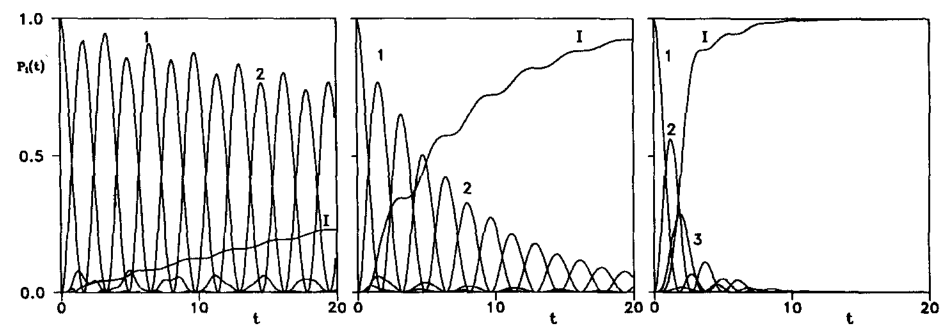


(*a*) (*b*) (*c*)

Figure 2. Time evolution of the level's population $P_i(t)$ and ionization of a resonantly excited four-level system with strong second Rabi frequency ($\Omega_1=1$, $\Omega_2=4$ and $\Omega_3=1$) for three loss rate values (*a*) $\gamma=0{\cdot}2$; (*b*) $\gamma=1$; (*c*) $\gamma=16$.

than the two external fields $\Omega_2 \gg \Omega_1$, Ω_3, the population is concentrated on the ground and the final level. The time evolution of the level's population of these two different schemes of an effective two-level systems is displayed for three various values of γ in figures 2 and 3.

The first case (figure 2) when $\Omega_2 \gg \Omega_1 \Omega_3$ is analogous to the case demonstrated by CSG in [2] (in both cases the effective two-level system is realized between the ground and the uppermost level from which a decay is allowed). Therefore it must be expected that the rate constants derived by CSG correctly describe the time evolution of population probabilities presented in figure 2. Indeed, for a decay rate γ much smaller than all Rabi frequencies and smaller than the effective Rabi frequency ($\Omega_{\text{eff}}=\Omega_1\Omega_3/\Omega_2$) the ground-state population decays nearly exponentially at a rate γ (figure 2 (*a*)). In figure 2 (*b*), in which γ is greater than Ω_{eff} but less than Ω_2, the population damps at a reduced rate $\Omega_{\text{eff}}^2/\gamma$. However, when γ becomes much larger than Ω_2 (figure 2(*c*)) the four-level atom behaves more like a damped



(*a*) (*b*) (*c*)

Figure 3. The same as in figure 2, but for the case of a strong third Rabi frequency ($\Omega_1=1$, $\Omega_2=1$ and $\Omega_3=4$).

three-level system, and not as a damped two-level system. The latter is quite natural if one takes into account that the conditions for validity of the two-level approximation are no longer valid for values of γ exceeding Ω_2 [4, 10]. We would like to point out in particular that in a regime of an effective two-level behaviour between the ground and the highest excited state, because the magnitude of the effective Rabi frequency is very small (an order smaller than any of the incident Rabi frequencies), fast ionization will be observed only for loss rate much smaller than any of the Rabi frequencies.

The completely opposite case arises when the third Rabi frequency is very strong and the system's population is localized between the ground and the second level. Now, as one can see from figure 3 (*a*), the addition of a small loss from the fourth level manifests itself as very slow damping in the oscillating population between the first and second levels. It is not difficult to derive that for values of γ less than Ω_3 the population decay rate is given by the expression $\gamma_{\text{eff}} = (\Omega_2/\Omega_3)^2\gamma$ (see appendix). Since this effective decay rate is much smaller than the corresponding effective Rabi frequency ($\gamma_{\text{eff}} < \Omega_1$, for $\gamma < \Omega_3$), the population decay becomes more pronounced with increasing γ (as is illustrated in figure 3 (*b*)). However, an increase in γ above $2\Omega_3$ leads to destruction of the condition for validity of the two-level approximation and part of the population reaches the third level. Again we observe in figure 3 (*c*) a reduction in the dynamics of the four-level system to that of a damped three-level system.

In figure 4, another two schemes of resonantly pumped four-level system are illustrated. In figure 4 (*A*) the lossless four-level system is prepared as an effective three-level atom (the third level is not populated) and in figure 4 (*B*) the population of the loss-free four-level atom is almost evenly distributed between all system levels.

Numerical experiments, presented in the above figures, clearly show some pronounced regularities in the time evolution of the population loss dynamics. First of all, we can see that the presence of losses much smaller than the Rabi frequencies ($\gamma \ll \Omega_i$) does not alter the excitation regime. Thus the population loss dynamics of the effective two-, three- or four-level atom continues to follow the dynamics of the respective lossless atom and the corresponding Rabi oscillations appear as exponentially declining oscillations. As long as the loss rate may be treated as low (when stimulated processes dominate dynamics), we should expect exponential enhancement of population damping with increasing γ. However, it is clearly seen that the value of the 'low' loss rate itself is very different for each concrete scheme of excitation. Losses which are small for the case illustrated in figure 3 are still large for the case in figure 2. Moreover, when the highest excited state remains unpopulated during the pumping process (as in figure 3 and presented below in figure 5), incorporation of a loss mechanism from this level produces population decay at rate radically different from γ and as a rule less than γ. (In the appendix, we shall demonstrate derivation of γ_{eff} for the two cases of effective two-level systems illustrated in figures 3 and 5.) When γ increases and the irreversible loss process successfully competes with the coherent processes, then most of the population decays during the first cycle (e.g. figure 4 (*b*)). There is always (for each arbitrary set of Ω_1 and D_1) a loss rate value for which the population damps most rapidly and hence the ionization occurs at the highest rate. An increase in γ above this optimum value leads to reduction in the ionization rate. When γ becomes much greater than any of the applied Rabi frequencies, the population loss dynamics strictly follow the dynamics of a three-level system, undergoing decay from the third level at a rate Ω_3^2/γ. Depending on the Ω_1 and Ω_2 magnitudes the population of this reduced three-level system will reside predominantly on the first state if Ω_2 exceeds Ω_1 by an order (figure 2 and figure 4 (*A*)); if $\Omega_1 > \Omega_2$, the population will be shared between first and second level (figure 4 (*B*))

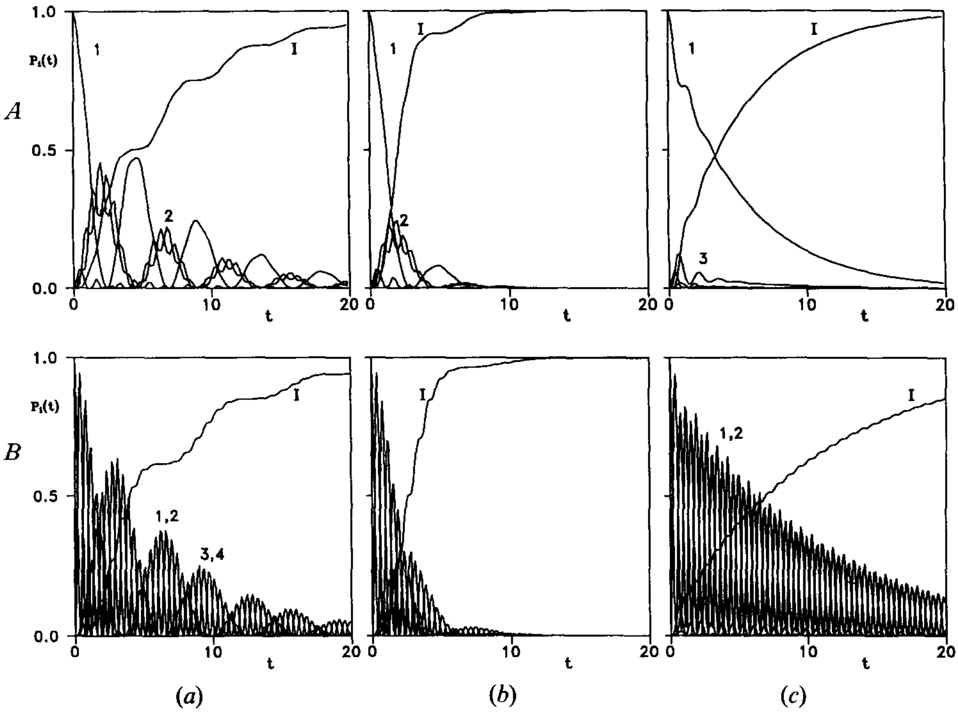


Figure 4. Population loss dynamics of two different regimes of a resonantly excited four-level atom for (*A*) $\Omega_1 = 1$, $\Omega_2 = 4$ and $\Omega_3 = 4$ and (*B*) $\Omega_1 = 4$, $\Omega_2 = 1$ and $\Omega_3 = 4$: (*a*) $\gamma = 0{\cdot}3$; (*b*) $\gamma = 1$; (*c*) $\gamma = 10$.

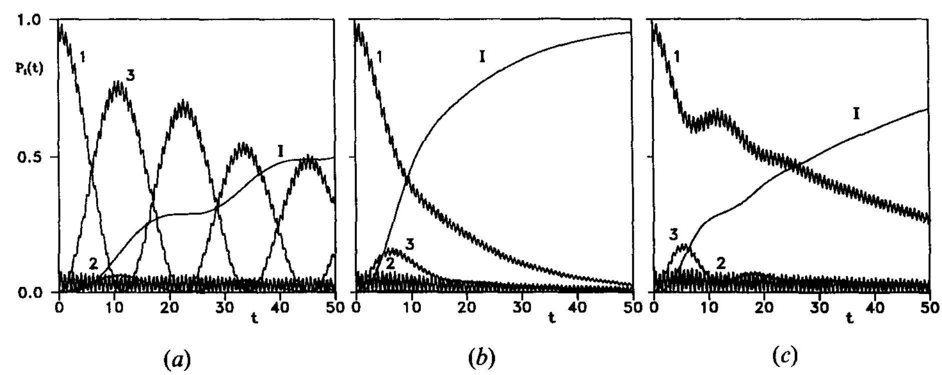


Figure 5. Time evolution of the population and ionization of a four-level system prepared as an effective two-level system between the first and third levels in the presence of γ, for $\Omega_1 = 1{\cdot}2$, $\Omega_2 = 1$, $\Omega_3 = 1{\cdot}5$, $D_1 = 8$, $D_2 = 0{\cdot}4$ and $D_3 = 5$: (*a*) $\gamma = 0{\cdot}2$; (*b*) $\gamma = 5$; (*c*) $\gamma = 16$.

and, for $\Omega_1 \approx \Omega_2$, the population will be distributed amongst the three levels (figure 3).

It is clear that the regularities stated above will be valid for the more general case of off-resonance excitation. It is evident that, in order to predict the history of population loss dynamics, one must know the relationships between Rabi frequencies and resonance detunings which determine one definite excitation regime. For illustration, in figure 5 we present the time evolution of the level's population of a four-level system pumped in a regime of effective two-level behaviour between the first and third level. Such behaviour occurs at large cumulative detunings D_1 and D_3 [10]. This example is similar to that shown in figure 3, in the sense that in both cases the uppermost state is not populated during the course of pumping. As is seen, for small γ the population of the effective two-level atom exponentially damps at a rate given by the expression $\gamma_{\mathrm{eff}} = \gamma\Omega_3^2/(D_3^2+\gamma^2)$ (see equation (A 7)). With increase in γ the ionization rate increases and, for value of γ higher than all the dynamic parameters (figure 5 (c)) the population loss dynamics again approach those of a slowly damped three-level system.

3.2. *Theoretical analysis*

Here we shall derive analytically the conditions under which the uppermost level decouples from the rest of the levels, as well as the dynamic parameters characterizing the resulting three-level system.

For this purpose we apply a specific procedure based on time-independent perturbation theory [11]. We shall restrict ourselves to exact resonance excitation.

Let us rewrite equation (1) in the following block form:

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C_a \\ C_b \end{bmatrix} = \begin{bmatrix} H_a & V \\ V^{+} & -\mathrm{i}\gamma \end{bmatrix}\begin{bmatrix} C_a \\ C_b \end{bmatrix}, \tag{2}$$

where subsystem a consists of levels 1, 2, 3 and subsystem b of level 4. The two groups of levels are coupled to each other by the operator V containing the Rabi frequency Ω_3.

The question arises: when it is possible to consider V as a small ('perturbative') correction to the 'zeroth-order' block-diagonal Hamiltonian

$$\mathbf{H}_0 = \begin{bmatrix} H_a & 0 \\ 0 & H_b \end{bmatrix}$$

describing levels 1–4 as two uncoupled subsystems a and b. To resolve this question we start by diagonalizing $\mathbf{H}_0$, using a block-diagonal unitary matrix

$$\mathbf{U} = \begin{bmatrix} U_a & 0 \\ 0 & 1 \end{bmatrix}.$$

Here $\mathbf{U}_a$ is a 3×3 unitary matrix, diagonalizing $\mathbf{H}_a$. It is not difficult to build $\mathbf{U}_a$ from the normalized eigenvectors of $\mathbf{H}_a$ in the following explicit form [12]:

$$\mathbf{U}_a = \begin{bmatrix} \Omega_2/G & \Omega_1/2^{1/2}G & \Omega_1/2^{1/2}G \\ 0 & -1/2^{1/2} & 1/2^{1/2} \\ -\Omega_1/G & \Omega_2/2^{1/2}G & \Omega_2/2^{1/2}G \end{bmatrix}, \tag{3}$$

where $G = (\Omega_1^2+\Omega_2^2)^{1/2}$.

The unitary transformation of equation (2) yields

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C'_1 \\ C'_2 \\ C'_3 \\ C'_4 \end{bmatrix} = \begin{bmatrix} \lambda_1 & 0 & 0 & V'_{14} \\ 0 & \lambda_2 & 0 & V'_{24} \\ 0 & 0 & \lambda_3 & V'_{34} \\ V'_{41} & V'_{42} & V'_{43} & \lambda_4 \end{bmatrix} \begin{bmatrix} C'_1 \\ C'_2 \\ C'_3 \\ C'_4 \end{bmatrix}, \tag{4}$$

where $\lambda_1 = 0$, $\lambda_{2,3} = \pm G$ are the eigenvalues of $\mathbf{H}_a$ (these are the usual 'dressed-atom' states of a resonantly driven three-level atom) and $\lambda_4 = -\mathrm{i}\gamma$.

The transformed components of the operator V have the form

$$V' = \mathbf{U}_a^{-1} V = \begin{bmatrix} -\Omega_1\Omega_3/2G \\ \Omega_2\Omega_3/2^{3/2}G \\ \Omega_2\Omega_3/2^{3/2}G \end{bmatrix}. \tag{5}$$

Now that the 'zeroth-order' Hamiltonian is diagonal, we are able to write down the well known conditions for applicability of the perturbation approach, namely the matrix elements of the perturbative operator must be small compared with the corresponding differences between the unperturbed eigenvalues:

$$|V'_{a4}| \ll |\lambda_a - \lambda_4|, \qquad \text{for } a = 1, 2, 3. \tag{6}$$

For the problem considered here, these conditions attain the following explicit form:

$$\frac{\Omega_1\Omega_3}{G} \ll \gamma; \qquad \frac{\Omega_2\Omega_3}{2^{1/2}G} \ll (G^2 + \gamma^2)^{1/2}. \tag{6 a}$$

It is readily seen that conditions (6 a) are satisfied when

(1) $\gamma \gg G$, $\gamma \gg \Omega_3$, that is the loss rate γ is much greater than all the applied Rabi frequencies (which is the weak-field limit) and
(2) $G \gg \gamma \gg \Omega_3$, that is when one or both of Ω_1 and Ω_2 is/are much larger than Ω_3, and γ exceeds only Ω_3 (case of a weak third field).

Below we shall determine the explicit form of the reduced effective Hamiltonians for these two cases and shall show that they describe different temporal behaviours of the reduced three-level system.

Applying the standard technique of perturbation theory, we find the transformed probability amplitudes with the first-order corrections in the form:

$$C_1'^{(1)} = \begin{bmatrix} 1 \\ 0 \\ 0 \\ V'_{41}/\lambda_1 - \lambda_4 \end{bmatrix}, C_2'^{(1)} = \begin{bmatrix} 0 \\ 1 \\ 0 \\ V'_{42}/\lambda_2 - \lambda_4 \end{bmatrix}, C_3'^{(1)} = \begin{bmatrix} 0 \\ 0 \\ 1 \\ V'_{43}/\lambda_3 - \lambda_4 \end{bmatrix},$$

$$C_4'^{(1)} = \begin{bmatrix} V'_{14}/\lambda_4 - \lambda_1 \\ V'_{24}/\lambda_4 - \lambda_2 \\ V'_{34}/\lambda_4 - \lambda_3 \\ 1 \end{bmatrix}.$$

Now, equation (4) expressed on the basis of these amplitudes can be symbolically written in the form

$$
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C_a'^{(1)} \\ \\ C_4'^{(1)} \end{bmatrix} \begin{bmatrix} \lambda_a - \dfrac{|V'_{a4}|^2}{\lambda_4-\lambda_a} + \lambda_a \dfrac{|V'_{a4}|^2}{(\lambda_4-\lambda_a)^2} & \dfrac{V'_{a4}|^2}{\lambda_4-\lambda_a}\sum \dfrac{|V'_{a4}|^2}{\lambda_4-\lambda_a} \\ \dfrac{V'_{a4}}{\lambda_4-\lambda_a}\sum \dfrac{|V'_{a4}|^2}{\lambda_4-\lambda_a} & \lambda_4 + 2\sum \dfrac{|V'_{a4}|^2}{\lambda_4-\lambda_a} \end{bmatrix} \begin{bmatrix} C_a'^{(1)} \\ \\ C_4'^{(1)} \end{bmatrix}. \tag{7}
$$

Equations (7) show that up to terms of second order the subsystems a and b may be considered as independent. The evolution of each subsystem can be derived by separating equations (7) into two sets: one for C'_a ($a=1, 2, 3$) and the second for C'_4.

Let us consider first the case of a high loss rate ($\gamma \gg \Omega_i$). When third-order terms are neglected in equation (7), the relevant equations of motion for amplitudes C'_a can be written in the form

$$
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C'_1 \\ C'_2 \\ C'_3 \end{bmatrix} = \begin{bmatrix} -\dfrac{|V'_{14}|^2}{\lambda_4} & -\dfrac{V'_{14}V'_{24}}{\lambda_4} & -\dfrac{V'_{14}V'_{34}}{\lambda_4} \\ -\dfrac{V'_{14}V'_{24}}{\lambda_4} & \lambda_2 - \dfrac{|V'_{24}|^2}{\lambda_4} & -\dfrac{V'_{24}V'_{34}}{\lambda_4} \\ -\dfrac{V'_{14}V'_{34}}{\lambda_4} & -\dfrac{V'_{24}V'_{34}}{\lambda_4} & \lambda_3 - \dfrac{|V'_{34}|^2}{\lambda_4} \end{bmatrix} \begin{bmatrix} C'_1 \\ C'_2 \\ C'_3 \end{bmatrix}. \tag{8}
$$

Now, in order to go back to the initial basis states C_a, we carry out the inverse unitary transformation on equation (7). In this way we obtain

$$
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix} = \begin{bmatrix} 0 & \Omega_1 & 0 \\ \Omega_1 & 0 & \Omega_2 \\ 0 & \Omega_2 & -\mathrm{i}\Omega_3^2/\gamma \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix}. \tag{9}
$$

Thus the dynamics of a four-level system with decay γ much greater than the largest of the Rabi frequencies is reduced to those of a three-level system with losses from the third level at rate Ω_3^2/γ. Population histories displayed in figures 2 (c), 3 (c), 4 (A) (c), 4 (B) (c) and 5 (c) are perfectly described by this effective 3×3 Hamiltonian.

Let us now turn to the second case when γ exceeds only Ω_3. On applicaton of an exactly similar procedure, as in the former case, the following set of equations for the probability amplitudes C_a for the case of strong first and second Rabi frequencies, is obtained:

$$
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix} = \begin{bmatrix} \mathrm{i}\Omega_3^2/4\gamma & \Omega_1 & \mathrm{i}\Omega_3^2/4\gamma \\ \Omega_1 & 0 & \Omega_2 \\ \mathrm{i}\Omega_3^2/4\gamma & \Omega_2 & -3\mathrm{i}\Omega_3^2/4\gamma \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix}. \tag{10}
$$

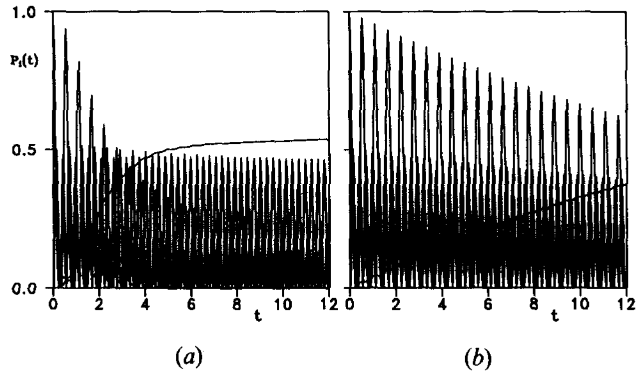


Figure 6. Reduced population loss dynamics of a four-level system to dynamics of a damped three-level system: (*a*) $\Omega_3 < \gamma \ll G$, for $\Omega_1 = \Omega_2 = 8$, $\Omega_3 = 1$ and $\gamma = 2{\cdot}5$; (*b*) $\gamma > \Omega_3$, G, for $\Omega_1 = \Omega_2 = 8$, $\Omega_3 = 1$, and $\gamma = 16$.

As one can see, in this case, simultaneously with the induced widths of the levels, there appear non-zero off-diagonal elements corresponding to direct coupling of levels 1 and 3. This result was unexpected and non-trivial to us and that is why we shall present the solutions of equation (10) in explicit form.

For the particular case $\Omega_1 = \Omega_2 = \Omega > \gamma > \Omega_3$, the solutions of equation (10) can be expressed (for a time interval which is not very long) approximately as

$$P_1(t) = \tfrac{1}{8}[1 + \cos(2^{3/2}\Omega t)] + \tfrac{1}{4}\exp\left(-\frac{\Omega_3^2}{\gamma}\right)t + \tfrac{1}{2}\cos(2^{1/2}\Omega t)\exp\left(-\frac{\Omega_3^2}{2\gamma}\right)t,$$

$$P_2(t) = \tfrac{1}{4}[1 - \cos(2^{3/2}\Omega t)],$$

$$P_3(t) = \tfrac{1}{8}[1 + \cos(2^{3/2}\Omega t)] + \tfrac{1}{4}\exp\left(-\frac{\Omega_3^2}{\gamma}\right)t - \tfrac{1}{2}\cos(2^{1/2}\Omega t)\exp\left(-\frac{\Omega_3^2}{2\gamma}\right)t,$$

$$I(t) = \tfrac{1}{2}\left[1 - \exp\left(-\frac{\Omega_3^2}{\gamma}\right)t\right].$$

These expressions show that the ionization probability increases until it is saturated at $\tfrac{1}{2}$ and the population of the second level remains insensitive to losses (figure 6 (*a*)). An inverse population is created in the channel 2–1. When γ becomes comparable with Ω the off-diagonal coupling between levels 1 and 3 can be neglected completely and equations (10) are transformed into equation (9).

4. Summary

We have presented numerical as well as analytical solutions of the population loss dynamics of a strongly excited four-level system with allowed decay out of the system from the highest state. Our aim has been to show that population loss dynamics essentially depend upon the relationships between the Rabi frequencies and resonance detunings. These relationships determine the behaviour of the original lossless four-level system to be as that of an effective two-, three- or four-level quantum system. It has been demonstrated how, on the basis of knowledge of these relations, we are able to give not only a qualitative but also a quantitative picture of the temporal behaviour of population dynamics and to explain the so-called unexpected (at first glance) results.

The results obtained can obviously be regarded as generalization of some results published earlier on the population loss dynamics of three-level systems [8]. On the other hand, these results can be generalized to N-level atoms, strongly driven by $N-1$ lasers.

In general, the observed regularities can be summarized as follows. The presence of a small loss from the uppermost level does not alter the excitation regime and population loss dynamics are closely related to those of coherently driven lossless multilevel atom. So long as stimulated processes dominate dynamics, the familiar Rabi oscillations damp almost exponentially at a rate γ when the ionization process is going from the final level, or at an effective loss rate when the uppermost level is not directly involved in the ionization process. There is always a loss rate value for which the decay proceeds most rapidly. An increase in γ above this optimum value leads to reduction in the population decay rate. For a loss rate greater than all dynamic parameters, the dynamics of the N-level system reduce to those of an $(N-1)$-level system with induced decay out of the system at a rate Ω_{N-1}^2/γ.

Finally, we would like once again to note that, if the N-level system is prepared as an effective two-level system between the ground and the highest level, from which the loss is added, because of the very small value of the effective Rabi frequency a significant fraction of ionization can be expected only for a loss rate several orders smaller than any of the applied Rabi frequencies. Otherwise, if γ is not sufficiently small, the system as a whole will remain insensitive to the incident laser fields, as well as to the force causing the losses.

Acknowledgment

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Appendix

In this appendix we shall derive the expressions for the effective decay rates for the cases demonstrated in figure 3 and figure 5.

We follow the formalism developed by Shore and Cook [4] for extracting two-level behaviour from a coherently driven N-level system.

Localization of the entire system population within two states only is mathematically equivalent to reducing the set of equations (1) to two coupled equations for probability amplitudes $C_1(t)$, $C_k(t)$, for $k=2, 3, 4$, which have the general form

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C_1 \\ C_k \end{bmatrix} = \begin{bmatrix} 0 & \Omega_{\text{eff}} \\ \Omega_{\text{eff}} & \Delta_{\text{eff}} \end{bmatrix} \begin{bmatrix} C_1 \\ C_k \end{bmatrix}, \tag{A 1}$$

where Ω_{eff} and Δ_{eff} are the effective two-level Rabi frequency and the effective resonance detuning respectively.

The Shore-Cook method, applied to a coherently driven four-level system, leads to an analytically soluble problem for arbitrary strengths and detunings of the incident fields. The conditions for validity of two-level behaviour, as well as the exact expressions for Ω_{eff} and Δ_{eff} for each of the level pairs $1-k$ (for $k=2$, 3, 4), have been determined in [10]. Here we shall use the expressions from [10], formally including the negative imaginary part to D_3.

Firstly, when the population is localized between the ground and the second level, equation (A 1) takes the form

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} C_1 \\ C_2 \end{bmatrix} = \begin{bmatrix} 0 & \Omega_1 \\ \Omega_1 & \Delta_{\mathrm{eff}} \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \end{bmatrix}. \tag{A 2}$$

The effective Rabi frequency coincides with the first Rabi frequency and the effective resonance detuning is

$$\Delta_{\mathrm{eff}} = D_1 - \Omega_2^2 \frac{D_3}{D_2 D_3 - \Omega_3^2}. \tag{A 3}$$

The condition for validity of two-level approximation is

$$|\tfrac{1}{2}\Delta_{\mathrm{eff}} \pm [(\tfrac{1}{2}\Delta_{\mathrm{eff}})^2 + \Omega_1^2]^{1/2}| \ll | -\tfrac{1}{2}(D_2 - D_3) \pm [\tfrac{1}{4}(D_2 - D_3)^2 + \Omega_3^2]^{1/2}|. \tag{A 4}$$

Substituting D_3 by $D_3 - \mathrm{i}\gamma$, for the exact resonance case displayed in figure 3, equations (A 3) and (A 4) become

$$\Delta_{\mathrm{eff}} = -\mathrm{i}\gamma \left(\frac{\Omega_2}{\Omega_3}\right)^2 \equiv -\mathrm{i}\gamma_{\mathrm{eff}}, \tag{A 5}$$

$$| -\mathrm{i}\tfrac{1}{2}\gamma_{\mathrm{eff}} \pm (-\tfrac{1}{4}\gamma_{\mathrm{eff}}^2 + \Omega_1^2)^{1/2}| \ll |\mathrm{i}\tfrac{1}{2}\gamma \pm (-\tfrac{1}{4}\gamma^2 + \Omega_3^2)^{1/2}|. \tag{A 6}$$

Equations (A 5) and (A 6) show that the resonantly excited four-level system with allowed decay at a rate γ from the highest level and the third Rabi frequency much greater than the first behaves as a damped effective two-level system with induced decay from the second level at rate $\gamma_{\mathrm{eff}} = \gamma(\Omega_2/\Omega_3)^2$ as long as $\gamma < 2\Omega_3$. For values of γ exceeding $2\Omega_3$ the condition (A 6) is violated and the system departs from the effective two-level behaviour. Now the conditions (6) are satisfied and the temporal evolution of the system is governed by the set of equation (9).

Secondly, in the limit of a two-level approximation between the first and third levels the probability amplitudes C_1 and C_3 satisfy equations (A 1) for $k=3$ with effective Rabi frequency $\Omega_{\mathrm{eff}} = \Omega_1\Omega_2/D_1$ and effective detuning

$$\Delta_{\mathrm{eff}} = D_2 + \frac{\Omega_1^2 - \Omega_2^2}{D_1} - \frac{\Omega_3^2}{D_3}\left(1 - \frac{\gamma^2}{D_3^2 + \gamma^2}\right) - \mathrm{i}\frac{\Omega_3^2\gamma}{D_3^2 + \gamma^2}. \tag{A 7}$$

The condition for validity of two-level behaviour here is

$$|\tfrac{1}{2}\Delta_{\mathrm{eff}} \pm [(\tfrac{1}{2}\Delta_{\mathrm{eff}})^2 + \Omega_{\mathrm{eff}}^2]^{1/2}| \ll |D_1|,\ |D_3|. \tag{A 8}$$

As one can see, in this case, the presence of γ introduces not only a negative imaginary part but also a real part to Δ_{eff}. For small γ ($\gamma \ll D_3$) the real part can be neglected and only the presence of the effective decay rate modifies the temporal behaviour of the effective two-level system, causing damping at a rate $\gamma_{\mathrm{eff}} \approx \gamma(\Omega_3/D_3)^2$. For values of $\gamma \geqslant D_3$, Δ_{eff} becomes larger than Ω_{eff}, that is the condition for effective population of the upper level in a two-level atom is not satisfied and the population occupies mainly the ground state. We see that for values of γ exceeding all the dynamic parameters the condition (A 8) remains valid and at the same time the conditions (6) are also valid. Therefore the population time evolution is equally well described by the set of equations (A 1) with Δ_{eff} given by equation (A 7), as well as by the set of equations (9) with non-zero diagonal matrix elements D_1 and $D_2 - i\Omega_3^2/\gamma$ respectively.

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