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Dynamics of multilevel laser excitation: Three-level atoms*

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We display the time dependence of populations in coherently excited three-level atoms. In particular we examine the relationship between ion production and the dynamical parameters: Rabi frequencies, detunings, and loss rate.

I. INTRODUCTION

The model two-level atom, driven near-resonantly by a single laser, has been studied extensively.[1] Details of the dynamical behavior have been reported in numerous articles and monographs, and a considerable corpus of intuition is now available for understanding such models. Much current interest attaches to such processes because of potential application to isotopically selective excitation, ionization, dissociation, or reaction.

Generalization to multilevel excitation, though introducing no essentially new physics, does require extension of our intuition regarding two-level processes to admit the increased degrees of freedom and enlarged parameter space.[2] Although it is possible to obtain analytic expressions for special cases, notably when parameters tend to extreme values, thereby permitting simple approximations, the general cases become too complicated for simple analytic treatment and one is led eventually to numerical computation. By relying at the outset on numerical rather than analytic approaches, one readily treats intense fields acting for long times and so accesses a large portion of parameter space. When displayed graphically, the computations often admit simple interpretation. In the following note we present illustrative examples of the dynamical behavior of multilevel atoms, pointing out some of the regularities.

Figure 1 symbolizes the model system we shall consider: a succession of atomic energy levels E_1, E_2, E_3 driven by lasers of intensities I_1 and I_2 and angular frequencies ω_1 and ω_2. We assume the lasers to be nearly monochromatic, to be turned on abruptly at $t=0$, and to be tuned near the appropriate Bohr frequency, $\hbar\omega_n \cong E_{n+1}-E_n$. We assume the laser intensities to be sufficiently great that stimulated emission and absorption processes dominate the dynamics; we neglect spontaneous decay and collision-induced relaxation. Only the irreversible loss of probability from the uppermost level at rate γ, referred to as ionization loss, disturbs the laser-driven dynamics.

Our model exemplifies situations wherein the dynamics is controlled by coherent stimulated processes. When incoherent processes, such as spontaneous decay, collisional interruption, or laser incoherence, become the dominant rates, then one must employ a more general formulation such as that provided by the density matrix or the Bloch vector (cf. Brewer and Hahn[3]).

Our interest centers on the time dependence of probabilities $P_i(t)=|C_i(t)|^2$ for finding the atom in level i, given initial certainty of the ground level, $P_1(0)=1$. The complex probability amplitudes $C_i(t)$ satisfy the time-dependent multilevel rotating-wave-approximation (RWA) Schrödinger equation[4]:

$$i\frac{\partial}{\partial t}C_i(t)=\sum_j W_{ij}C_j(t)\,, \tag{1}$$

where, for three-level excitation, the effective non-Hermitian time-evolution operator W has the form

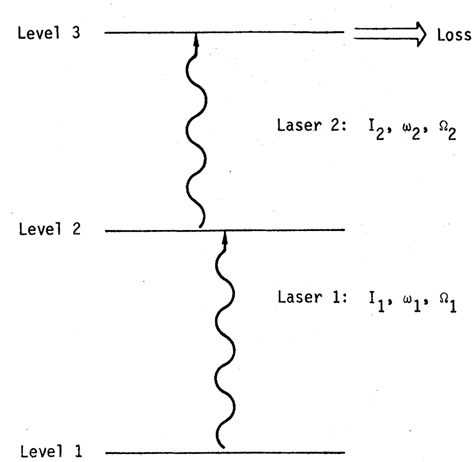

FIG. 1. Model three-level atom.

$$W=\begin{pmatrix}\Delta_0 & \frac{1}{2}\Omega_1 & 0\\ \frac{1}{2}\Omega_1 & \Delta_1 & \frac{1}{2}\Omega_2\\ 0 & \frac{1}{2}\Omega_2 & \Delta_2-\frac{1}{2}i\gamma\end{pmatrix}. \tag{2}$$

(For N-level excitation W becomes an $N\times N$ matrix, still of tridiagonal form with non-Hermiticity still confined to element W_{NN}.)

The RWA utilizes the following parameters: The loss (decay) rate γ, related to the imaginary part of the effective Hamiltonian $H^{\rm eff}$ or to the lifetime τ,

$$\hbar\gamma=-2\,{\rm Im}(H_{33}^{\rm eff})=\hbar/\tau\;; \tag{3}$$

the detuning Δ_n of n successive laser frequencies away from the nth Bohr combination frequency,

$$\begin{aligned}\hbar\Delta_n&=0,\quad n=0\\ &=E_{n+1}-E_1-\hbar(\omega_1+\omega_2+\cdots+\omega_n),\quad n\geq 1;\end{aligned} \tag{4}$$

and the (resonant) *Rabi frequency* Ω_n equal to the peak instantaneous value of the interaction Hamiltonian or to

$$\hbar\Omega_n=|H_{n,n+1}|_{\rm max}=(8\pi I_n/c)^{1/2}d_{n,n+1}, \tag{5}$$

where d is the component of the transition dipole moment along the electric field.

In the following sections we shall examine the effects on solutions $P_i(t)$ of varying the loss rate γ, the Rabi frequencies Ω_1 and Ω_2, and finally the detunings Δ_1 and Δ_2.

II. VARYING LOSS-RATE γ

In the absence of ionization loss, probability is conserved and merely cycles through the three levels. With both lasers tuned to resonance (zero detuning Δ_i) and adjusted in intensity for equal Rabi frequency, the probability oscillations are as shown in Fig. 2(a). These oscillations, obviously generalizations of the two-level Rabi oscillations, have been published by Sargent and Horowitz.[5] Population flows from level 1 through level 2 to level 3 and returns. In this resonant excitation the dominant populations are those at the top and bottom of the excitation ladder; the intermediate-level population oscillates more rapidly and with smaller amplitude.

It should be noted that the simple periodicity of Fig. 2(a) reflects the occurrence of but a single nonzero eigenfrequency in the 3×3 rotating-wave Hamiltonian matrix; in general (i.e., with more levels) the eigenfrequencies are incommensurable and the population oscillations do not exhibit any simple periodicity.[6]

The addition of a small loss mechanism from level 3 damps the oscillations as shown in Fig. 2(b). As in the case with two levels, the total

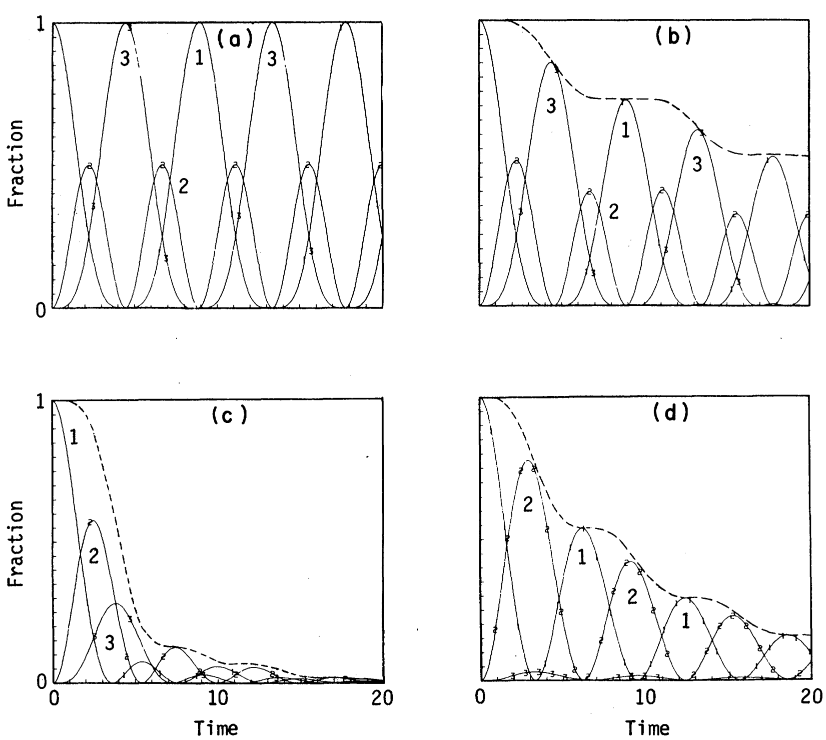

FIG. 2. Fractional populations as a function of time showing effect of loss rate γ upon each level of a three-level atom: (a) $\gamma=0$, (b) $\gamma=0.1$, (c) $\gamma=1$, (d) $\gamma=5$. All plots have equal Rabi frequencies $\Omega_1=\Omega_2=1$ and run for times $0\leq t\leq 20$.

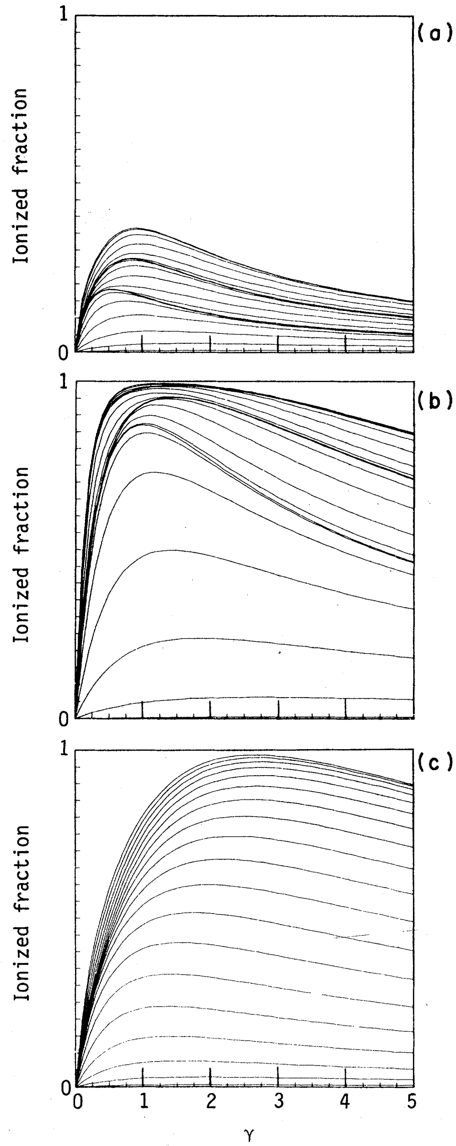

FIG. 3. Fractional ionization as a function of loss rate γ. Curves correspond to successively later time, separated by increments $\Delta t = 1$: (a) $\Omega_1 = 1.0$, $\Omega_2 = 0.3$; (b) $\Omega_1 = \Omega_2 = 1.0$; (c) $\Omega_1 = 0.3$, $\Omega_2 = 1.0$.

bound-state fraction (the sum of populations in levels 1, 2, and 3) follows a modulated exponential decline. Figure 2(b) shows this bound fraction as a dashed curve enveloping the population curves. The steplike declines, a prominent general feature of multilevel excitation and ionization, coincide with population growth of the ionizing level 3; the ionization rate varies in direct proportion to the probability amplitude of level 3.

By increasing the loss rate γ out of level 3 until it approximates the Rabi frequencies, we can speed the loss of bound probability; i.e., we increase the effective ionization rate.

Figure 2(c) shows the population oscillations for this case: two resonant lasers whose equal Rabi frequencies equals the loss rate. We see here a dramatic loss of probability with the first cycle through level 3; some 85% of the atoms ionize during this cycle, with nearly all the remaining atoms following during the next few cycles.

By further increasing the loss rate γ we diminish the population in level 3, thereby reversing the aforementioned trend and slowing the effective ionization rate. Figure 2(d) shows the population oscillations when the loss rate γ is 5 times the two equal Rabi rates; the ionization rate and the level-3 populations are both substantially smaller here than in the previous figure. As the loss rate γ is made still larger, the dynamics eventually approach that of an undamped two-level system.

The preceding figures imply that, for fixed Rabi frequencies (i.e., fixed excitation-laser intensity), there is an optimum loss rate γ which will maximize the ionization within a given time interval. Figure 3(b) displays this relationship by plotting, for a sequence of times, the total ionized fraction as a function of the loss rate γ. For comparison, Fig. 3(a) and 3(c) present plots for unequal Rabi frequencies. These and similar computations show that, although precise values depend upon the time interval as well as upon relative Rabi frequencies, as a rule of thumb *for multilevel resonant excitation with nearly equal Rabi frequencies, ionization occurs most rapidly when the loss rate γ is approximately equal to the Rabi rate Ω.* When γ is smaller or larger than Ω (by a factor exceeding 2), the ionization proceeds appreciably slower.

This rule applies only to situations in which excitation progresses along a single unbranched excitation route to a final ionizing level; the occurrence of ionization from another level in the chain can alter very dramatically the dynamics, as we shall discuss in a future publication.

III. VARYING RABI FREQUENCIES Ω

The examples of Fig. 2 dealt with equal Rabi frequencies and no detuning. For small loss rates ($\gamma \lesssim \Omega$) the probability flows cyclically from level 1 through level 2 on to level 3 where it reverses and returns to level 1. This pattern changes when the second-step laser weakens in intensity, as illustrated in Fig. 4(a). Here the second-step Rabi frequency is 0.3 times the first-step value (both are tuned resonantly to the Bohr frequency) so that, after population flows from level 1 into level 2, a portion returns to level 1 rather than pro-

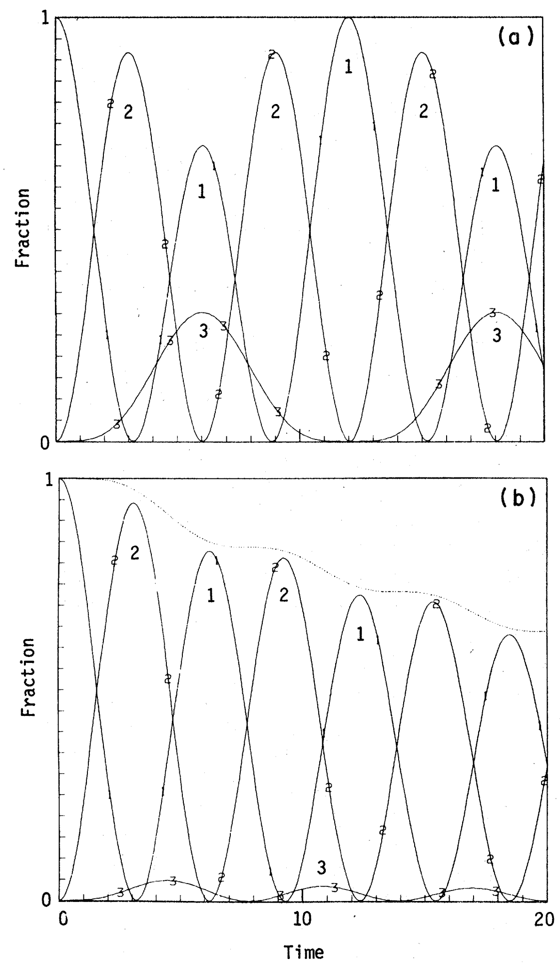

FIG. 4. Populations of three-level atom as a function of time, for $0 \le t \le 20$, when Rabi frequencies are $\Omega_1 = 1.0$, $\Omega_2 = 0.3$: (a) $\gamma = 0$, (b) $\gamma = 1.0$.

ceeding up the excitation ladder. The pattern is just what we would expect for a two-level atom (levels 1 and 2) weakly coupled into a third level: two-level Rabi oscillation weakly modulated by the coupling to level 3.

This two-level pattern persists in the presence of loss, as we see in Fig. 4(b). As with equal Rabi rates, there is an optimum γ for most rapid ionization: with γ too small there is no loss, with γ too large the population of level 3 remains small. Figure 3(a) shows that, although the optimum varies with time, it is never far from Ω_1 (unity here).

The time behavior appears qualitatively different if the first excitation step has the weaker laser. Figure 5(a) reverses the order of the previous Rabi frequencies, making the first-step frequency 0.3 times the second, still with both lasers resonantly tuned to their respective Bohr frequencies.

[Because we neglect spontaneous emission there is no difference between the dynamics of excitation and that of deexcitation. Figure 5(a) could equally well refer to an initially populated excited state, labeled 1, weakly coupled to a two-level atom. Only the initial conditions and the labeling of states differ in the dynamics depicted in Figs. 4(a) and 5(a).]

The probability flow here, as in the case of equal Rabi frequencies, is through level 2 into level 3; there is no back-reflection of probability flow as there is when flow occurs toward smaller Rabi frequencies. However the most striking feature of Fig. 5(a) is the incomplete modulation of levels 1 and 3: the initially populated level 1 is never completely depopulated.

The pattern of incomplete modulation occurs whenever Rabi frequencies successively increase

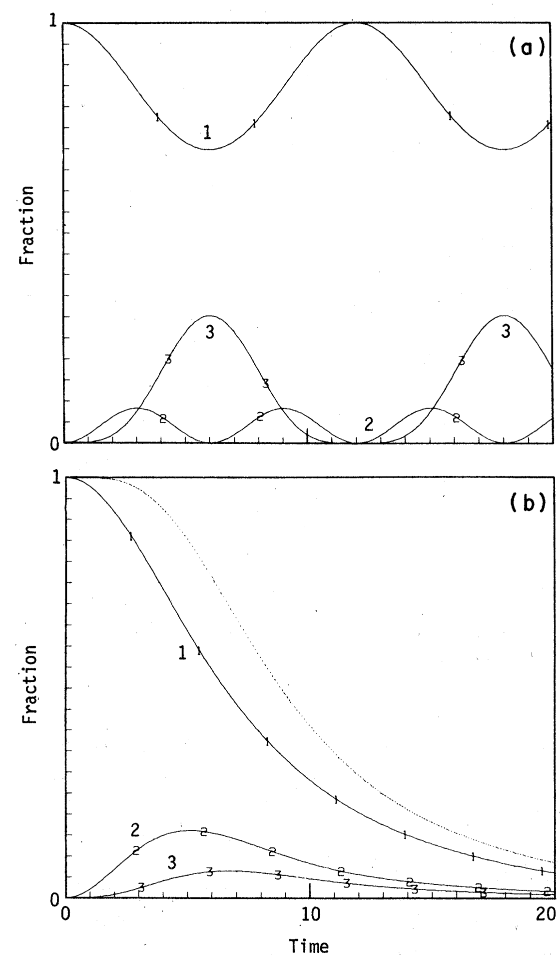

FIG. 5. Populations of three-level atom, for $0 \le t \le 20$, when Rabi frequencies are $\Omega_1 = 0.3$, $\Omega_2 = 1.0$: (a) $\gamma = 0$, (b) $\gamma = 1.5$.

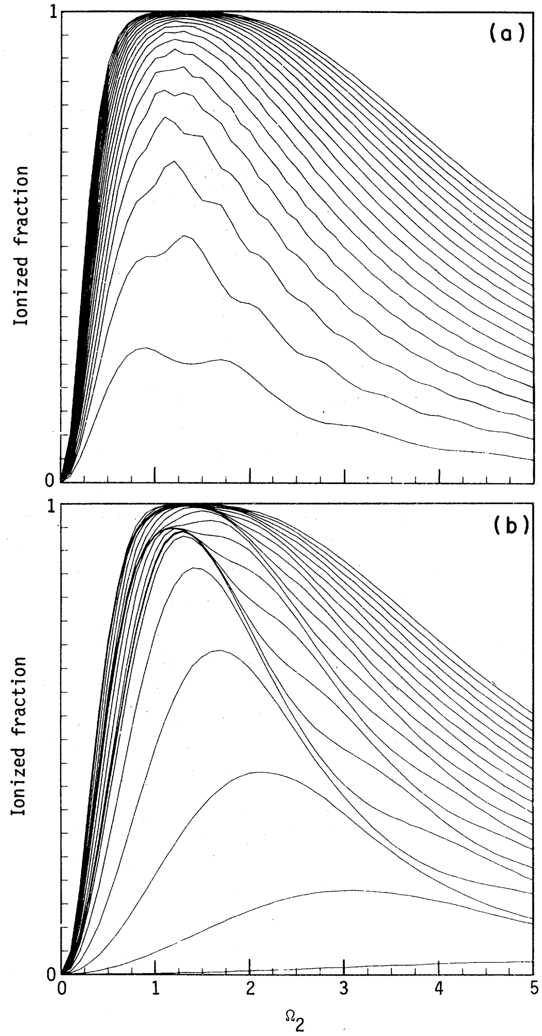

FIG. 6. Fractional ionization of three-level atom for various times as a function of $0 \leq \Omega_2 \leq 5.0$ for $\Omega_1 = 1$: (a) $\gamma = 0.1$, successive curves separated by $\Delta t = 10$; (b) $\gamma = 1$, successive curves separated by $\Delta t = 1$.

as one proceeds up the excitation ladder, with all lasers tuned to the Bohr frequencies. It also persists in the presence of ionization loss. We shall comment further on this in the next section.

In all of the curves hitherto displayed, the populations undergo oscillations. It is interesting to find situations where only a single pulsation occurs during the course of ionization. Figure 5(b) depicts such a case for the three-level atom. Population flows steadily out of level 1, through level 2, into level 3 from which it ionizes. Such critically damped solutions are well known for the two-level atom, and they can be found for N-level systems. They occur for a range of Rabi frequencies which are adjusted to steadily increase at each higher step along the excitation ladder.

From the preceding graphs one expects that, for fixed detuning and fixed decay rate, ionization will proceed most rapidly when successive Rabi frequencies increase. Figure 6 validates this expectation: we see here the ionization as a function of the second Rabi frequency for a succession of times and for loss rates $\gamma = 0.1$ and $\gamma = 1$. Although precise optimization depends upon the chosen time interval and loss rate, we are not greatly in error by taking Ω_2 to be 25% larger than Ω_1.

The case of progressively increasing Rabi frequencies combined with a top-level loss rate γ greater than the largest Rabi frequency has been shown to be one of the few multilevel problems for which rate equations apply no matter how intense and coherent the exciting laser fields are.[7] The nonoscillatory populations of Fig. 5(b) illustrate this rate-equation behavior.

IV. VARYING DETUNING

The occurrence of incomplete depopulation, as illustrated in Fig. 5 is familiar from studies of two-level atoms. Here, as there, the behavior is associated with detuning. For all of the preceding figures we set detunings to zero: the lasers are tuned to the free-atom atomic Bohr frequencies, as defined in the limit of negligible perturbation by the lasers. However, it is known that a weak field, probing the structure of a two-level atom driven by an intense field, reveals a doublet structure. The frequency separation grows in proportion to the rms value of the (resonant) Rabi frequency and the detuning. Thus, to obtain maximum probe response for the two-level atom and complete modulation of population oscillations, one must tune the weak probe not to the unperturbed Bohr frequency but to one of two shifted frequencies. This level splitting is the dynamical Stark effect.[8]

Figure 7(a) shows the effect of detuning the weaker first-excitation laser an amount equal to half the larger Rabi frequency. We see that the population leaves completely the initial level 1 and enters a Rabi-oscillating two-level system thereby increasing the overall amount of population reaching the ionizable level 3. Only for these two detunings ($\Delta = \pm\frac{1}{2}$) is there complete modulation of level 1. Levels 2 and 3 form a strongly coupled pair which oscillate about a time-dependent average population. Figure 7(b) shows the effect of ionization loss in this case.

These "dressed-atom" resonances are apparent when we scan in frequency the weaker of the two lasers.[9] Figures 8(a) and 8(b) show such scans. The peaks or maxima signal the occurrence of a

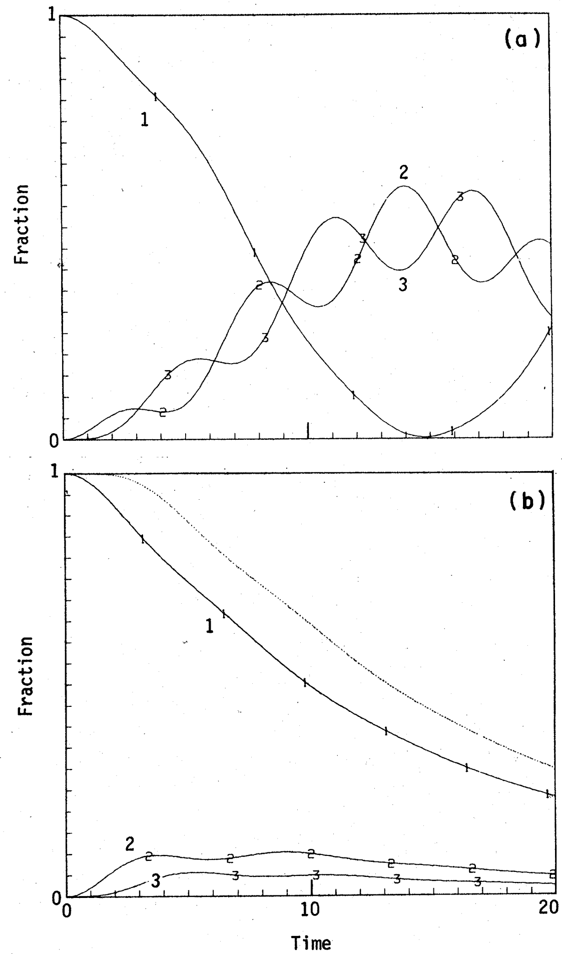

FIG. 7. Populations of three-level atom as a function of time, $0 \leq t \leq 20$, showing effect of detuning first level by $\Delta_1 = 0.5$, with Rabi frequencies $\Omega_1 = 0.3$, $\Omega_2 = 1.0$: (a) $\gamma = 0$, (b) $\gamma = 1$.

two-photon resonance.

If we scan in frequency the more intense of the two lasers as shown in Figs. 8(c) and 8(d), we see less evidence of dynamic Stark splitting. The small splitting of the weak laser, though perceptible at short times, is masked by the power broadening of the stronger laser. By increasing the detuning of the intense laser, we increase its induced Stark splitting. Consequently the two-photon transition never passes through a resonance.

V. DOPPLER DETUNING

In a vapor source the thermal motion of irradiated atoms introduces Doppler shifts to the laser light. We consider vapors for which phase-interrupting or velocity-changing collisions are infrequent, so that excitation remains coherent. Then the influence of thermal motion can be modeled by an ensemble average over Doppler detunings.

Consider first the case of collinear lasers—both lasers propagating in the same direction. For simplicity, let the two lasers have nearly equal frequencies, so that to a moving atom each laser frequency ω appears Doppler shifted by the same amount Δ. The operator W of Eq. (4) now has the special form

$$W = \begin{pmatrix} 0 & \frac{1}{2}\Omega_1 & 0 \\ \frac{1}{2}\Omega_1 & \Delta & \frac{1}{2}\Omega_2 \\ 0 & \frac{1}{2}\Omega_2 & 2\Delta - \frac{1}{2}i\gamma \end{pmatrix}. \tag{6}$$

Note that, because each laser is shifted by Δ, the two-photon energy deviates from resonance by 2Δ. Figure 9 shows the populations of each level as a function of time and detuning Δ, for Doppler shifts of collinear lasers. We observe that as detuning increases the populations of level 1 and 2 oscillate more rapidly. This phenomena is well-known in the two-level atom: populations pulsate at a frequency equal to the rms value of detuning and resonant Rabi frequency. Further we observe that ionization is appreciable only for a range of detunings comparable to the Rabi frequency; for larger detunings the population never reaches the third level.

Consider next the case of counter-propagating lasers—lasers propagating in opposite directions. Now the Doppler shift of one laser, Δ, is offset by an opposite shift, $-\Delta$, for the other laser. As a result, the matrix W is

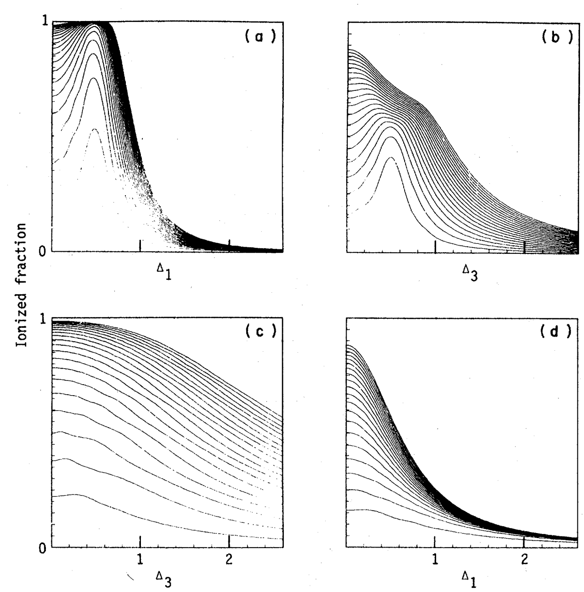

FIG. 8. Fractional ionization of three-level atom as a function of detunings Δ_1 or Δ_2 for $\gamma = 0.1$: (a) $\Omega_1 = 0.3$, $\Omega_2 = 1$, vary Δ_1; (b) $\Omega_1 = 1.0$, $\Omega_2 = 0.3$, vary Δ_2; (c) $\Omega_1 = 0.3$, $\Omega_2 = 1.0$, vary Δ_2; (d) $\Omega_1 = 1.0$, $\Omega_2 = 0.3$, vary Δ_1.

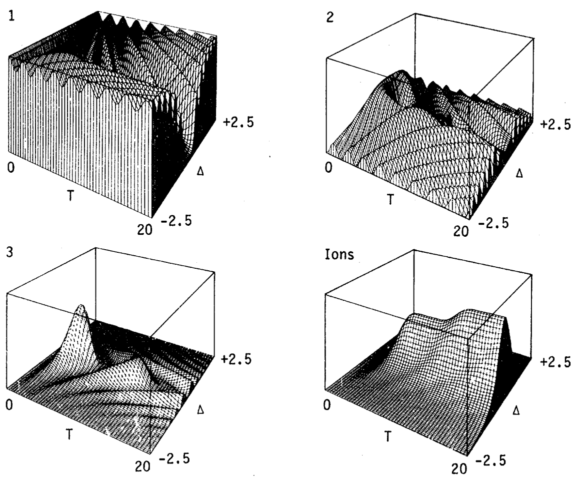

FIG. 9. For collinear lasers, the separate frames show populations in levels 1, 2, and 3 and the ions as a function of time T and as a function of the first-transition detuning Δ (in units of the equal Rabi frequencies). The cumulative detunings are $\Delta_1=\Delta$ and $\Delta_2=2\Delta$. Exact resonance (no detuning) is represented by a vertical plane bisecting the Δ axis and parallel to the T axis; an instantaneous snapshot of population distributions is given by a vertical plane perpendicular to the T axis.

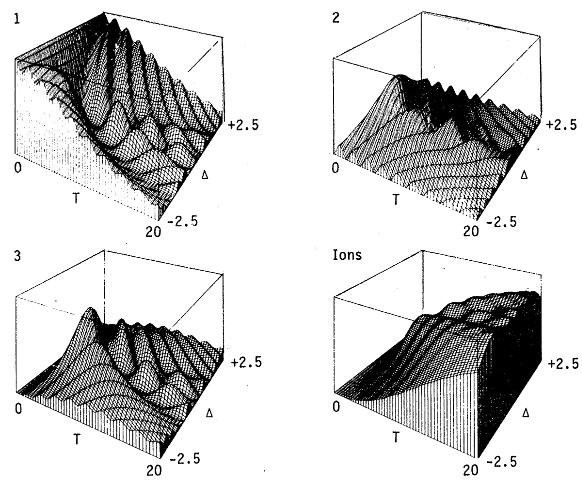

FIG. 10. For counter-propagating lasers (or for a virtual level), the separate frames show populations in levels 1, 2, and 3 and the ions as a function of time T and as a function of the first-transition detuning Δ (in units of the equal Rabi frequencies). The cumulative detunings are $\Delta_1=\Delta$ and $\Delta_2=0$.

$$W=\begin{pmatrix} 0 & \frac{1}{2}\Omega_1 & 0 \\ \frac{1}{2}\Omega_1 & \Delta & \frac{1}{2}\Omega_2 \\ 0 & \frac{1}{2}\Omega_2 & -\frac{1}{2}i\gamma \end{pmatrix}. \quad (7)$$

The two-photon transition is now resonant for all Doppler components—one has the familiar Doppler-free absorption. For those atoms which have large detunings, the second level is a "virtual" level: its population, never very large, oscillates at high frequency. This high-frequency modulation of populations in levels 1 and 3 is apparent in Fig. 10.

VI. SUMMARY

The behavior of a coherently driven three-level system is governed by the Rabi frequencies, detunings, and loss rate.

For fixed loss rate, ionization occurs most rapidly when the Rabi frequencies are comparable in magnitude; particularly favorable is the case where successive Rabi frequencies increase by some 25%. When Rabi frequencies are very different, the overall ionization rate is degraded. Some improvement in ionization can be gained by detuning the weaker lasers achieving an n-photon resonance.

For fixed and comparable Rabi frequencies, ionization occurs most rapidly when the loss rate is comparable to the Rabi rate.

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[1] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

[2] For our modeling of successive multistep laser excitations of atoms leading to ionization, multilevel excitation with lasers propagating in the same directions introduces essentially no new physics beyond the two-level atom. However, recently some new effects have been both observed and predicted for three-level atoms: For effects due to two-photon Stark shifts see M. M. T. Loy, Phys. Rev. Lett. 36, 1454 (1976) and references therein; for Doppler-free spectroscopy see G. Grynberg, F. Biraben, M. Bassini, and B. Cagnac, Phys. Rev. Lett. 37, 283 (1976) and references therein; R. Salomaa and S. Stenholm, J. Phys. B 8, 1795 (1975).

[3] R. G. Brewer, E. L. Hahn, Phys. Rev. A 11, 1641 (1975).

[4] The form of the RWA time-dependent Schrödinger equation used here is well known; for example, see M. P. Silverman and F. M. Pipkin, J. Phys. B 5, 1844 (1972); I. M. Beterov and V. P. Chebotaev, *Three-Level Gas Systems and Their Interaction with Radiation* (Pergamon, New York, 1974), see Sec. II.

[5] M. Sargent and P. Horowitz, Phys. Rev. A 13, 1962 (1976). For further analytic and numerical examination of N-level excitation see J. H. Eberly, B. W. Shore, Z. Bialynicka-Birula, and I. Bialynicki-Birula (to be published).

[6]Although there are known examples of nearly periodic behavior for n-level atoms (e.g., the truncated harmonic oscillator), the solutions for the choice $\Omega_1 = \Omega_2 = \cdots = \Omega_n$ generally show no simple periodicities for $n > 3$. An exception is level 3 of the five-level atom.

[7]J. R. Ackerhalt and J. H. Eberly, Phys. Rev. A 14, 1705 (1976).

[8]E. T. Jaynes and F. W. Cummings, Proc. IEEE 51, 89 (1963); C. R. Stroud, Jr., Phys. Rev. A 3, 1044 (1971).

[9]J. L. Picqué and J. Pinard, J. Phys. B 9, L77 (1976) and references therein.