Coherent dynamics of N-level atoms and molecules. I. Numerical experiments*

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We study numerically the solution to Schrödinger's equation for the radiative interaction of fictitious N-level model atomic and molecular systems with monochromatic radiation fields. The numerical method of solution allows us to study multiphoton transitions in N-level systems unhampered by the assumptions usually adopted, such as weak fields or large detunings of intermediate states, in perturbative and quasiperturbative analytic studies. We point out regularities and near regularities in the population dynamics that appear to be qualitatively different from those encountered in two- and three-level systems.

I. INTRODUCTION

The two-level atom is one of the best known and most fruitful of atom models in optical resonance physics.[1] Multiple-level atomic and molecular systems have been of interest in quantum optics during the past several years because tunable-laser excitation of such systems allows the experimental study of various phenomena for the first time. Among these phenomena are such effects as resonant multiphoton ionization[2] and dissociation,[3] Doppler-free two-photon absorption,[4] isotopically selective multiphoton ionization[5] and dissociation,[6] strong-field resonance fluorescence,[7] and others.

It has become important to understand the dynamics of N-level atomic and molecular systems that are near-resonantly coupled to monochromatic radiation fields. Theoretical work in this area to date is almost completely confined to one of two approaches: the N-level system is thought of either as a sequence of connected two-level transitions, each stimulated by single-photon absorption, or as one giant two-level system whose single transition is stimulated by the simultaneous absorption of N -1 photons. The view of an N-level system as a sequence of two-level systems has its roots in the familiar rate-equation treatment of multistep processes, a treatment in which there is supposed to be no way to reach the mth level without first going through the $(m-1)$th. It was probably the pioneering work of Bebb and Gold[8] that served to emphasize the opposite possibility: that all of the levels between 1 and N could be discarded "completely," under certain far-from-resonance conditions,

thus permitting the N-level system to be viewed as a single two-level system whose excitation required $N-1$ photons simultaneously. This latter viewpoint has been developed extensively recently by Takatsuji[9] and other workers[10] to permit a simplified two-level treatment for detailed dynamics of resonantly interacting three-level atoms.

Neither of these two approaches is particularly well suited to the problems of recent interest. This is because near-resonance excitation is desired, not avoided, and because the electric field strengths and degree of monochromaticity commonly encountered in recent work make coherent and nonperturbative analyses imperative. The simplest generalization of the familiar two-level system is of course the three-level system. Recently, Sargent and Horowitz[11] displayed an exact and explicit solution (in the rotating-wave approximation) to the Schrödinger equation for a three-level atom with the assumption that none of the three levels are subject to decay. They also indicate how to modify their solution when all three levels are characterized by the same decay constant. They illustrate their results with a graph of the time dependence of the three levels'occupation probabilities when the lasers are adjusted so that the two transitions are being driven exactly on resonance and at the same Rabi rate.[12]

It is, of course, possible to solve the Schrödinger equation analytically in more general situations than the one they discuss, in particular, for atoms with more than three energy levels, irradiated by more than two monochromatic radiation fields, because the time-dependent ampli-

tude equations remain linear first-order differential equations no matter how many levels are considered, or what damping constants are assumed. This is certainly well-known, but the solution is not often attempted, because the diagonalization of an N-dimensional matrix is required for the N-level solution.

Closely related questions whose answers do *not* seem to be well known are: What is the highest dimensionality N for which an explicit *analytic* evaluation of the diagonalization problem can be accomplished? Can it be done off-resonance? Can it be done with different Rabi rates? With what kind of off-resonance detunings can it be done? More generally, one can simply ask whether or not there is a framework sufficiently broad to allow all of these questions to be discussed analytically at the same time. The important central question underlying all of these is: Just what is N-level atom behavior, and is it distinct from or roughly the same as two- and three-level behavior?

One of the motivations for undertaking an investigation of these questions arises from high-power laser experiments on atomic ionization, molecular dissociation, and chemical reactions. A general goal in these experiments is to attain as large a transition rate to an nth excited level as possible. It is not known in the general n-transition and m-laser case what arrangement of laser powers and detunings will lead to the highest rate, although some preliminary results are known. Ackerhalt and Eberly[13] have established the conditions under which multistep ionization and dissociation processes can be faithfully treated by purely rate-equation analysis, even in the high-power limit, without important error; and Garrison and Wong[14] have shown in two-level and three-level examples[12] that these conditions do not lead to the highest rates. The highest rates appear always to arise when a substantial degree of atomic coherence is present. Feldman and Elliott[15] have come to a similar conclusion in their study of multiphoton absorption by a 10-level model molecule.

Apart from possible application to multiphoton excitation, the problem of coherent monochromatic excitation of an N-level system is of intrinsic interest as one of the few soluble problems of quantum dynamics. Like the two-level atom and the harmonic oscillator, it provides insights into behavior of aspects of real systems.

Thus we here consider the problem of an N-level system coupled to monochromatic lasers that are nearly or exactly resonant with the $N-1$ intermediate transitions. We assume that the N near-resonance levels are the actual energy levels of the system, with all power-dependent shifts due to the existence of the remaining off-resonance levels al-

ready accounted for. We ignore the existence of any relaxation or damping processes. That is, we take the extreme view that the atom-field interaction is completely coherent. In this case all rate-equation approximations will fail, of course. The temporal behavior of the level-occupation probabilities can be expected to be very complicated, with the common feature being population pulsations rather than monotonic population flow.

The levels near the bottom of the excitation ladder and those near the top are particularly interesting. The probability is initially confined to the lowest levels, and we assume that the highest levels in the model correspond to those in a real atom or molecule where a useful laser-independent interaction or reaction occurs. Our completely coherent lossless model is relevant whenever this reaction (induced by, for example, static fields or collisions) occurs at a rate significantly smaller than the probability pulsation rate for the Nth level. Such an assumption can be relevant to multiple-photon atomic ionization if the cross section for the ionizing step is sufficiently low compared to that for the next lower discrete-level transition. The applicability to multiple-photon molecular dissociation processes is less clear because such processes are still poorly understood. However, if multiple-photon dissociation proceeds via a weak transition from a low-lying single-vibrational mode discrete-level ladder into a mixed-mode quasicontinuum, then our results should be useful in that context also.

In the following section we define our problem mathematically by stating the relevant form of the Schrödinger equation. In Sec. III we show graphs of several families of numerical solutions to the equation, choosing particularly regular relations among the dipole moments and field strengths appropriate to the various transitions. Among the interesting features of these numerical solutions are long-term quasiperiodic swings in occupation probabilities, surges of population into a certain "highest" level, and a nearly linear relation between the number of levels and the time required for the last level to first become substantially populated. In Sec. IV we present a first-order interpretation of a set of numerical "experiments." We point out broad features of the numerical solutions for the level populations, particularly features that have no theoretical formulas to support them. Finally, in Sec. V we summarize our findings, and suggest problems remaining for numerical study. In the accompanying paper we undertake an analytic examination of the examples explored numerically here, and develop a polynomial method of analysis applicable to a wide variety of N-level absorption problems.

II. EQUATIONS OF MOTION

Consider an N-level system with $N-1$ allowed consecutive transitions, as shown in Fig. 1. We imagine that one or several monochromatic lasers are present, so that all of the $N-1$ transitions can be stimulated at, or very near to, resonance. We assume that the individual laser frequencies ω_m are sufficiently close to the Bohr frequencies (the actual power-shifted transition frequencies of the system) $\Delta E_m/\hbar$ to justify a global rotating-wave approximation (RWA)[16] for the N-level system. Under this assumption, the Hamiltonian matrix becomes time-independent and tridiagonal, with zeros everywhere except along the two diagonals located one above and one below the main diagonal. That is, in a representation which has suitably chosen phases and which diagonalizes the N-level field-free Hamiltonian, the RWA Schrödinger equation for the transition amplitude $T_{mn}(t)$ to find the atom in state $|m\rangle$ at time t if it was initially in state $|n\rangle$ at $t=0$ is

$$i\frac{\partial}{\partial t}T_{m,n} = \tfrac{1}{2}\Omega_{m-1}T_{m-1,n} + D_{m-1}T_{m,n} + \tfrac{1}{2}\Omega_m T_{m+1,n} \quad (1)$$

with the initial condition

$$T_{mn}(t=0) = \delta_{mn} . \quad (2)$$

As Eq. (1) shows, the dynamics is governed by two families of parameters. The first is exemplified by

$$D_m = \sum_{k=1}^{m}(\omega_k - \Delta E_k/\hbar) + D_0 , \quad (3)$$

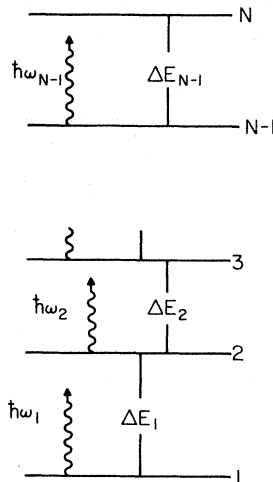the accumulated detuning at the mth transition. (D_0 is an arbitrary constant.) The other parameters,

FIG. 1. Notation appropriate to the transition of an N-level system.

the Rabi frequencies Ω_m, given by

$$\hbar\Omega_m = |\vec{d}_m \cdot \vec{\mathscr{E}}_m| , \quad (4)$$

are proportional to the peak values of the interaction energies of the dipole transition moments $\vec{d}_m$ (between levels m and $m+1$) and the electric field vectors $\vec{E}_m(t)$ (the field of the laser responsible for the mth transition):

$$\vec{E}_m(t) = \vec{\mathscr{E}}_m\cos(\omega_m t). \quad (5)$$

In the simple two-level situation the Rabi frequency has two interpretations: it is also the frequency of population oscillations between the two levels as well as equaling the interaction energy $|\vec{d}\cdot\vec{\mathscr{E}}|$ divided by $\hbar$. In the N-level case, as we will see, these two frequencies have no simple relationship. In the remainder of this paper we adopt the latter meaning for the term Rabi frequency.

Well-known techniques exist for computing numerically the solutions of coupled linear equations, of which the time-dependent Schrödinger equation (1) is a simple example. A particularly convenient method, applicable when the equation coefficients are time dependent, as they are in the RWA, makes use of the eigenvalues and eigenvectors of the coefficient matrix.

Various standard routines exist for the computation of eigenvalues and eigenvectors of given matrices. With their use it is a simple matter to solve Eq. (1) numerically and compute transition amplitudes and probabilities for any time t. In the next section we demonstrate some of these solutions for two particular choices of Rabi frequencies, assuming in all cases that the population is in level 1 at $t=0$.

First, however, we point out again that it is useful to solve for the full set of Schrödinger amplitudes $T_{mn}(t)$ for $m, n = 1, \ldots, N$ only when the corresponding population rate equations (PRE's) are inadequate.[13] Thus we should exhibit graphs of the off-diagonal quantities such as $T_{m1}T_{m'1}$ so as to show most dramatically the differences between the solutions to the full Schrödinger equation and the much simpler PRE solutions.[13] From each initial state $|n\rangle$ we obtain N^2 such complex-valued quantities. To show all of them would be impractical. Therefore we show only N quantities

$$P_{m1}(t) = |T_{m1}(t)|^2 \quad (6)$$

representing the population in level m evolving from the ground level $m=1$. The existence of off-diagonal coherence in the N-level system will still be evident from the distinctly nonmonotonic behavior of these level populations.

III. SAMPLE SOLUTIONS

The RWA Schrödinger equation (1) contains far too many independent parameters (the Ω's and the D's) to make a completely general solution useful as soon as N is larger than 3 or 4, even if one could be obtained analytically. There are cases, however, in which the Ω's and the D's obey simple rules among themselves, and which have some special significance.

Two examples of a simple rule[17,18] are (1) the case in which all Rabi frequencies are equal (the equal-Rabi case), which is the direct generalization of both the original two-level problem and recent work on the three-level case,[11,12] and (2) the case in which all of the $N-1$ Bohr transition frequencies are equal: $\Delta E_1 = \Delta E_2 = \cdots = \Delta E_{N-1}$, and the Rabi frequencies increase as $\sqrt{m}$, as they would in a purely harmonic vibrational mode of a molecule (the harmonic-Rabi case).

$$\text{harmonic: } D_m \equiv 0, \quad \Omega_m = \sqrt{m}\,\Omega_1; \tag{7a}$$

$$\text{equal: } D_m \equiv 0, \quad \Omega_m = \Omega_1. \tag{7b}$$

One can easily verify that these cases admit exact analytic solutions for $N = \infty$ level sequences.[17] For the harmonic-Rabi case the transition proba-

bility is

$$P_{m,1}(t) = \frac{(\Omega_1 t)^{2m-2}}{(m-1)!}\exp[-(\Omega_1 t)^2], \tag{8a}$$

whereas for the equal-Rabi case the transition probability is

$$P_{m,1}(t) = [J_{m-1}(\Omega_1 t)]^2 \tag{8b}$$

where $J_m(x)$ is the Bessel function of order m. We show in the accompanying paper that the truncated (finite N) sequence also admits exact solutions for these two cases.

In the remainder of this section we show graphs of numerical solutions to the time-dependent Schrödinger equation for these two finite-N cases in the absence of detuning. We comment on the most obvious features of the solutions.

In the first series of graphs in Fig. 2 we show, on the same scale, the behavior of the occupational probabilities of the levels of 3, 4, 7, and 15-level systems starting from a common initial condition: at $t=0$ the system is in its ground state. All Rabi frequencies are taken to be equal to the first. All of the levels of the three-level atom exhibit perfectly periodic pulsations, with the period and amplitude of levels 1 and 3 being twice as great
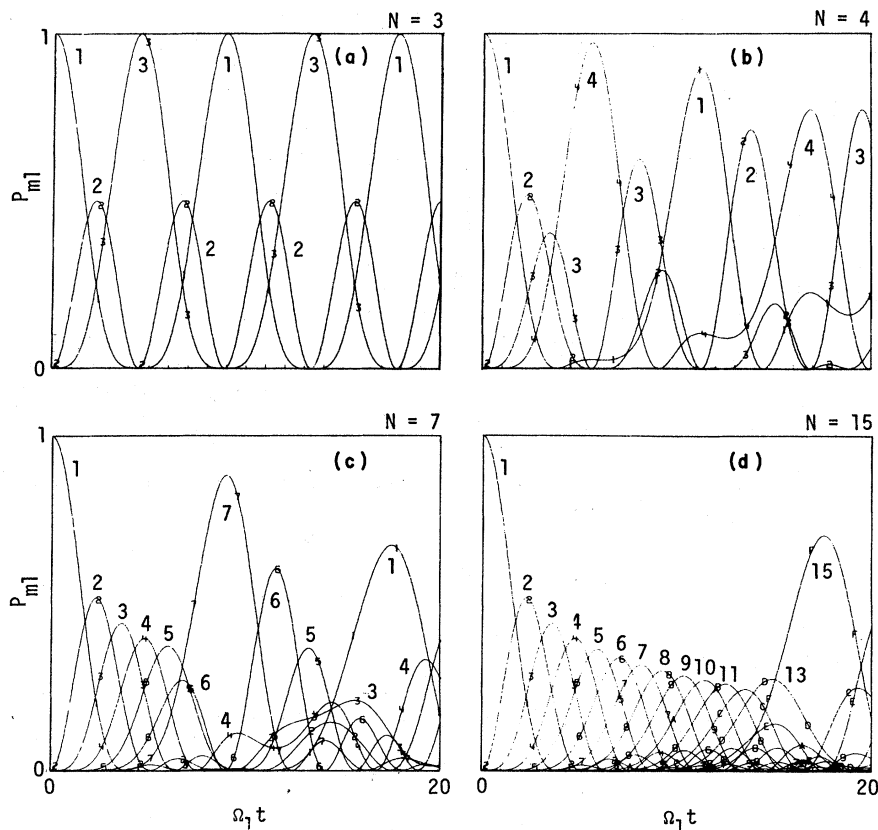

FIG. 2. Level populations $P_{m,1}(t)$ in the resonant ($D_m = 0$) equal-Rabi ($\Omega_m = \Omega_1$) case, as a function of $\Omega_1 t$, for $N = 3$, 4, 7, and 15 level systems.

as for level 2. However, the four-level example suggests that periodicities are absent in the general case.

The last two examples show an unexpected feature of the N-level system that appears to be general. After the population is driven by the laser radiation out of the first level, the population of each succeeding level rises to a peak and then subsides, with the populations peaking at lower and lower values, all of this with the notable exception of the final level. In each case the final level exhibits a significant surge in peak population compared with the lower levels immediately preceding it. This population surge might be explained pictorially by imagining the occupation probability as a fluid sloshing back and forth in a channel. As the fluid reaches either end of the channel, it accumulates to a greater height than at any intermediate position. The same behavior might also be interpreted as evidence that the excitation of the highest level does not occur entirely via a sequence of N steps, but also partially occurs via a direct $1 \rightarrow N$ channel. A direct multiphoton channel does not exist for a system described by incoherent population-rate equations, but could be expected to be implicit in the fully coherent Schrödinger equation.

Beers and Armstrong[19] have pointed out the possible importance of a direct channel for a realistic description of ionization.

The second series of graphs, in Fig. 3, shows the level populations in the N-level harmonic-Rabi case. The graphs in Fig. 3 exhibit features very similar to those shown for the equal-Rabi system in Fig. 2.

The complexity of the general N-level system, compared to the two-level and three-level systems, is apparent in the graphs of Figs. 2 and 3. Since the regularity of the Rabi oscillations in the smaller systems is such a pronounced feature of the dynamics, it is worth emphasizing the nonperiodicity of the corresponding oscillation in the general case. This is demonstrated in the graphs shown in Fig. 4. In these graphs the time behavior of the populations of each of the levels of five-level systems is shown. It is immediately clear that the middle level of the equal-Rabi five-level atom appears to oscillate perfectly periodically while none of the other levels appear to have more than a rough quasiperiodicity in their time behavior. The behavior of level 3 in this case appears to be perfectly periodic. This appearance is accurate, and is established analytically in the following paper.[18] It can
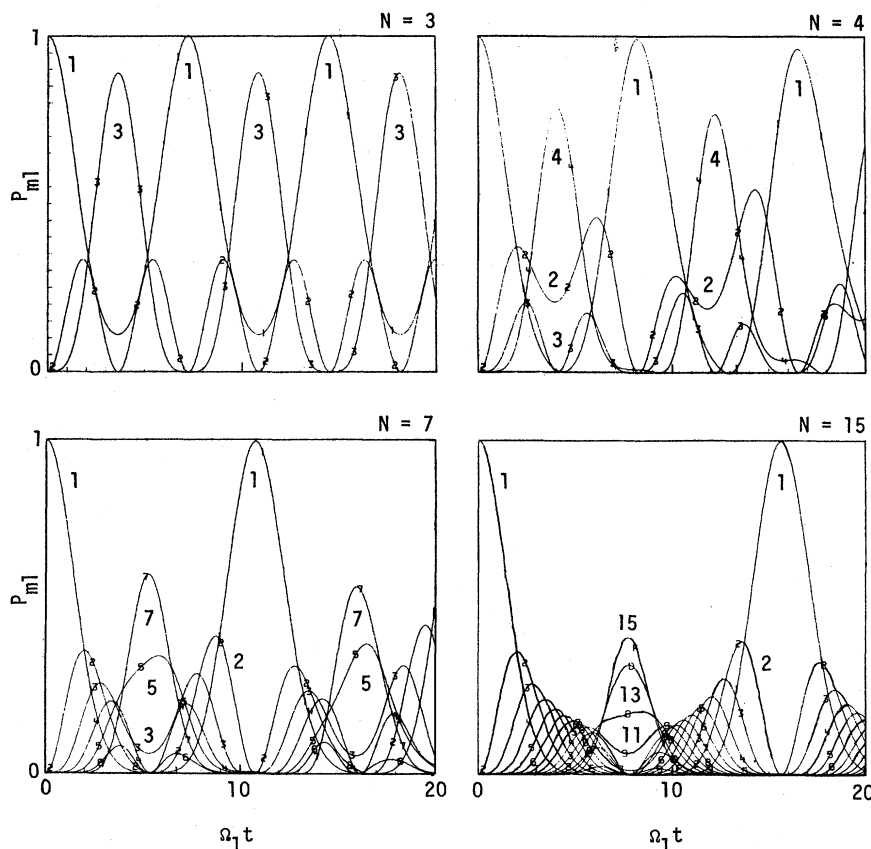

FIG. 3. Level populations $P_{m,1}(t)$ in the resonant ($D_m = 0$) harmonic-Rabi ($\Omega_m = \sqrt{m}\,\Omega_1$) case, as a function of $\Omega_1 t$, for $N = 3$, 4, 7, and 15 level systems.
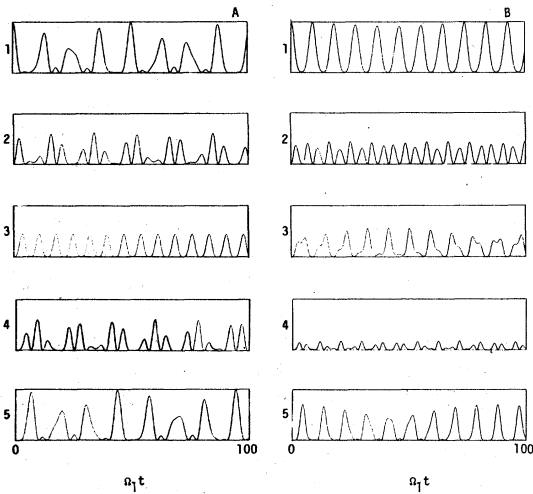
FIG. 4. Level populations $P_{m,1}(t)$ of a resonant $(D_m = 0)$ $N=5$ level system as a function of $\Omega_1 t$. A: equal-Rabi case $(\Omega_m = \Omega_1)$; B: harmonic-Rabi case $(\Omega_m = \sqrt{m}\,\Omega_1)$.

also be shown[18] that it is the only level population to vary periodically in any equal-Rabi atom larger than $N=3$. In order to emphasize this, we show in Fig. 5 the seven-level equal-Rabi atom. Note that neither the middle level, nor any of the others, shows periodic time behavior.

IV. THEORETICAL EXPERIMENTS IN N-LEVEL ATOMS

The strictly numerical *ab initio* integration of Schrödinger's equation is the most direct way to obtain reults for a specific system of interest. Numerical integration is less useful if one's main interest is in the qualitative properties of solutions for an entire class of problems. However, in
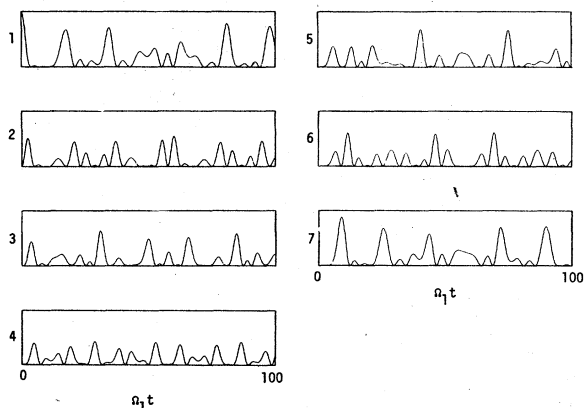

FIG. 5. Level populations $P_{m1}(t)$ of a resonant $(D_m = 0)$ $N=7$ level system as a function of $\Omega_1 t$ for an equal-Rabi $(\Omega_m = \Omega_1)$ sequence.

classes of problems of the kind considered in this paper, for which a simplified empirical or phenomenological theory has not yet been constructed, the numerical method can play a different, essentially "experimental," role. That is, the Schrödinger theory of the interaction of dipole transitions with radiation is certainly the correct basic theory, but much too complicated, as we have seen, when N is greater than 3, to permit quick and accurate semiquantitative predictions to be made. From the standpoint of our intuitive feel for the right results in a given circumstance, it is little better than no theory at all. Thus the result of every numerical integration can be regarded as the result of an experiment done to study the absorption of radiation by multilevel systems. And we can use these results to try to extract useful semiempirical truths that are not obviously revealed by the exact Schrödinger equation. Toward this goal, in this section we present some "experimental" results obtainable from our N-level graphs, and offer explanations for some of them.

The surge of population into the last level of an N-level system might be said to be expectable, because the probability "turns around" there before flowing back down into the lower levels. This does not explain another clear feature of the graphs, however. This feature is apparent in Fig. 3, but is much clearer in the long-time average populations shown in Fig. 6: namely, the anomalously low population *at all times* of the level immediately preceding the last one in a truncated oscillator. If the probability-fluid description is in-
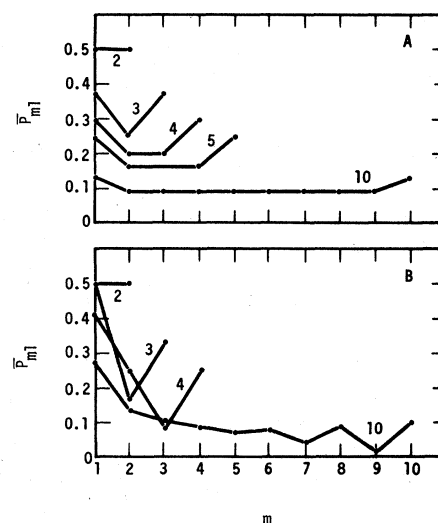

FIG. 6. Long-time average populations $\bar{P}_{m1}$ for several N-level systems; A: equal-Rabi case $(\Omega_m = \Omega_1)$ for $N = 2$, 3, 4, and 5 and 10; B: harmonic-Rabi case $(\Omega_m = \sqrt{m}\,\Omega_1)$, for $N=2$, 3, 4, and 10.
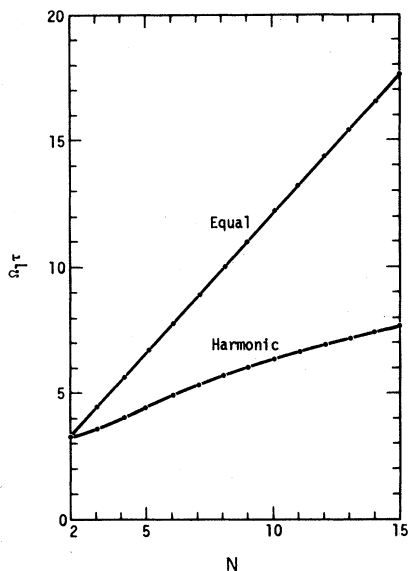
FIG. 7. Time to reach first maximum of population in Nth level of an N-level system, as a function of N, for harmonic-Rabi ($\Omega_m = \sqrt{m}\,\Omega_1$) case and for equal-Rabi ($\Omega_m = \Omega_1$) case.

FIG. 8. Maximum height of population curve for Nth level of an N-level system, during the initial population surge as a function of N, for harmonic-Rabi ($\Omega_m = \sqrt{m}\,\Omega_1$) case and for equal-Rabi ($\Omega_m = \Omega_1$) case.

voked here, the probability fluid seems to set up a standing wave near the top end of the excitation ladder, with the next to last level very near to a node.

Another feature of the population-dynamics graphs is shown in Fig. 7, where the time lapse before the occurrence of the surge in the last level is plotted as a function of the number of levels in the system, both for the equal-Rabi case and the harmonic-Rabi case. The equal-Rabi case is more striking because the time lapse appears to be very nearly a linear function of level number.

An almost equally regular feature of the population surge is the dependence of its maximum height on level number. As Fig. 8 shows, the maximum height falls off very smoothly, and rather slowly, as a function on N. The curves in Fig. 8 suggest the interesting possibility that there might be a universal asymptotic slope of the curve, common to all multilevel models. No theoretical confirmation or refutation of this possibility is known to us.

Another characterististic observable in numerical solutions, for which a heuristic explanation is possible, but which is also not directly predicted by any N-level-atom formula known to us, arises in the following way. Many molecules exhibit pronounced anharmonicity in their vibrational transition ladders, with the vibrational levels gradually getting closer together at higher vibrational quantum numbers. Consequently, a laser
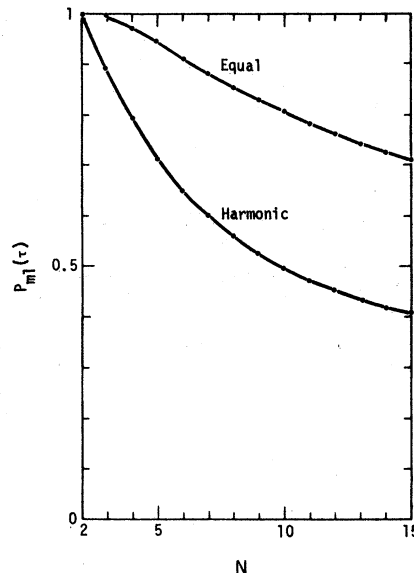
that is exactly resonant with the first vibrational transition will be detuned by an amount 2A, with A the anharmonicity parameter of the ladder, from two-photon resonance with the second transition, and will be detuned by an amount

$$D_m = m(m-1)A \tag{9}$$

from exact m-photon resonance with the mth transition.

In Fig. 9 we show the effect of such anharmonicity on the population dynamics of a ten-level harmonic Rabi system. In the first graph, for zero anharmonicity, we see behavior of the same kind shown in Fig. 3. That is, population flows to the top of the ladder, building up an appreciable amplitude in the last level, and avoiding the next-to-last level almost completely. Given a small anharmonicity, the graph, Fig. 9(b), shows that the behavior is roughly the same, with two differences: it is now the ninth level that is effectively the last, and there is some population found in the next-to-last eighth level. When the anharmonicity is made even larger, Fig. 9(c), the trend persists. Now the fifth level is effectively the last, and the fourth level is not avoided at all. Finally, the last graph shows the logical conclusion of such a trend. The anharmonicity has become so large that even the second transition is so far out of resonance that almost all of the population is confined to the first transition and simply swings back and forth between levels 1 and 2.
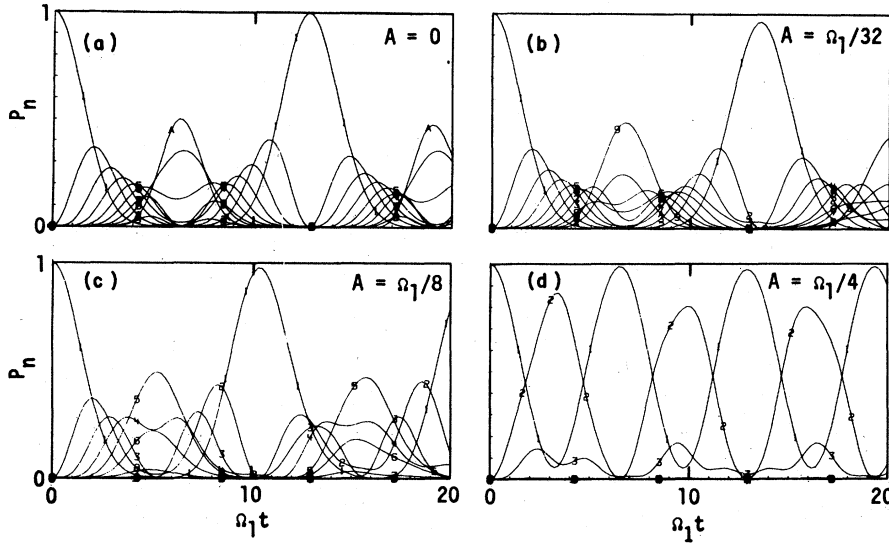
FIG. 9. Populations $P_{m1}(t)$ of levels of a 10-level harmonic-Rabi system $(\Omega_m = \sqrt{m}\,\Omega_1)$ with anharmonic detuning, $D_m = m(m-1)A$, as a function of $\Omega_1 t$ for anharmonicity $A = 0$, $\Omega_1/32$, $\Omega_1/8$, and $\Omega_1/4$.

A rough explanation for the trend shown in the graphs in Fig. 9 is easily found. It is natural that levels substantially out of resonance should receive little population. What is surprising is that there is always one level that acts as if it were the last. That is, population does not simply get smaller in levels farther and farther from resonance. Instead it surges into and "rebounds" from a certain detuned level whose probability equals or exceeds that of the levels immediately preceding it, and is much higher than that of any of the levels following it. We note that the level for which this occurs is predicted in a qualitative fashion by invoking the idea of ac-Stark broadening.[18] We can guess that the two-level ac-Stark effect will give each transition a "power broadened linewidth" related to its Rabi frequency. If this is so, then we can expect to induce transitions up the vibrational ladder until the accumulated detuning exceeds the Rabi frequency and not much further.[20]

Finally, we examine briefly a question that is especially important in the case of the N-level harmonic-Rabi system. We show in the following paper[18] that the truncated harmonic-Rabi system is exactly solvable. It remains true, however, that the untruncated harmonic oscillator is so simple in its dynamic behavior that it would be of only academic interest to study the truncated system if it were well approximated by the familiar untruncated one. Evidence that the N-level and the ∞-level oscillators are similar is already evident in Fig. 7. The time required for the population to flow into the highest level, shown in Fig. 7, is numerically very nearly proportional to the square root of the last level number. This is the behavior to be expected from the familiar ∞-level harmonic-oscillator transition-probability formula

(8a). That is, from (8a) one easily constructs the rule that the expected level number $\langle m(t) \rangle$ takes the form

$$\langle m(t) \rangle = \sum_{m=1}^{\infty} m P_{m,1}(t) = (\Omega_1 t)^2 + 1. \tag{10}$$

In other words, the time required for the mean level number to depart from $\langle m \rangle = 1$ is proportional to $(\langle m \rangle - 1)^{1/2}$.

A more thorough comparison of the truncated and untruncated oscillators is possible by using the well-known feature of the Poisson distribution (8a): the dispersion in level number is linearly related to the average level number. In the present case, where we have begun our level-number labeling at 1 instead of at 0, this means that

$$E(m) \equiv -\langle m \rangle - \langle m \rangle^2 + \langle m^2 \rangle + 1 = 0. \tag{11}$$

We show, in Fig. 10, a plot of $E(m)$ as a function of time for a 15-level truncated oscillator. For reference we also show the populations curves of the first and the 15th levels. It is clear that the truncated oscillator behaves very nearly like an untruncated one, with $E = 1$, for a major portion of the time required for the first population surge to flow into the top level. However, as soon as the 15th level begins to be populated the oscillator can recognize that it is not infinite in extent, and E departs significantly from unity. It is interesting to observe the extent to which the truncated oscillator can forget about the truncation at level 15, and return to the $E = 1$ mode of operation, as the population flows back down toward level 1. This might have been expected from the nearly perfect periodicity of the truncated oscillator curves in Fig. 3. From our point of view, the deviation from purely untruncated behavior near the
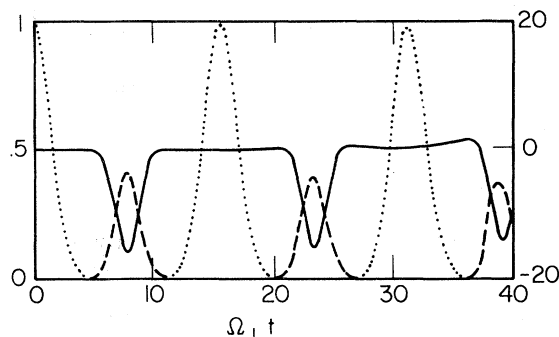
FIG. 10. Left-hand ordinate scale: dotted curve is population of level 1, $P_{1,1}(t)$; dashed curve is population of level 15, $P_{15,1}(t)$. Right-hand ordinate scale: solid curve is $E(t) = -\langle m \rangle - \langle m \rangle^2 + \langle m^2 \rangle + 1$.

top of the ladder is more important than the very nearly untruncated behavior in the lower levels because, of course, ionization or dissociation will occur near the top.

V. SUMMARY

In this paper we have explored numerically the solution to Schrödinger's equation for an N-level system near-resonantly coupled to electromagnetic radiation in the global rotating-wave and dipole approximations. Our interest has been in beginning to understand the N-level system in its own right, instead of as a sequence of two-level systems or as a single $(N-1)$-photon two-level system. It is these two limits that have been employed in most studies to date.

Our approach is also based on a simplified view of an N-level system. We have avoided approximations based on assumed small or large parameters, such as small electric field strength or large detunings from intermediate resonances, because it is just the regime of large field strengths and small detunings that is currently of most interest. We have instead considered only two, particularly "regular," kinds of N-level system: one in which all of the Rabi frequencies are equal, and one in which the Rabi frequencies obey the square root rule, $\Omega_m = \sqrt{m}\,\Omega_1$. These are not unrealistic N-level systems, although far from the most general. The equal-Rabi system can be realized in an atom simply by adjusting the various lasers' field strengths to compensate for variation in dipole moment from transition to transition. The so-called harmonic-Rabi system is well approximated by the vibrational ladder of a molecule. Letokhov and Makarov[17] have also treated exactly these two special cases, but from an approximate analytic approach.[18]

Our results are necessarily experimental, embodied for the most part in the graphs of level populations. We have observed a number of regular features that seem to be common to N-level systems, as soon as N is not 2 or 3, and this by itself may be the central result. That is, we have seen clearly that two-and three-level systems are almost singularly simple in their differences from general N-level behavior. Sargent and Horowitz[11] have reminded us that, for the most part, a three-level system is barely different from a two-level system. However, we see here that even a four-level system is qualitatively different from two-level and three-level systems, and closer in its nonperiodic population dynamics to a 10-level or 15-level system.

Our graphs suggest that it may be more than merely picturesque to think of probability as a fluid. In Figs. 2 and 3 we see behavior very similar to that expected of water in a channel with fixed ends. This appearance is heightened by an examination of time averages, and is discussed in the following paper.[18]

We see, particularly in Fig. 6, that the top few levels of a truncated N-level system are strikingly dissimilar in their temporal behavior: they are significantly distorted by the abrupt termination. Level N is overpopulated, and levels $N-1$, $N-3$, etc., are underpopulated, as compared with the extrapolation of preceding levels. This termination effect becomes increasingly pronounced with increasing N. By contrast, the effective termination caused by anharmonicity does not exhibit this enhancement/diminution effect. This means that models in which dissociation occurs only from the terminal level of an N-level system, as in the work of Mukamel and Jortner,[21] may give an unrealistic overestimate of dissociation. In any realistic molecular system, there will be no single level leading to dissociation. To our knowledge this feature of N-level dynamics has not yet been incorporated in an analytic or numerical treatment of photodissociation.

Another feature, the quasiperiods apparent in the graphs, for example the almost regular recurrence of substantial population of the initial level, is not explained by a direct examination of the atomic Hamiltonian. The frequency of these periods is much lower than any of the system's Rabi frequencies or Hamiltonian eigenvalues. They indicate strongly that the N-level system has an internal coherence, a coherence that permits interference and beat phenomena among the system's fundamental frequencies to become manifest even in the diagonal and "incoherent" variables, the level populations. These quasiperiods are also examined in the following paper.[18]

The internal coherence of the N-level system is one of its most intriguing features for further study. Its existence may suggest the existence of a valuable semiempirical theory underlying most of the obvious population dynamics, but containing far fewer parameters than the exact Schrödinger equation. To our knowledge no steps have yet been taken to delineate such a theory.

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