Coherent dynamics of N-level atoms and molecules. II. Analytic solutions*

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(Received 4 April 1977)

We show that a class of multiphoton-absorption problems has exact solutions. In these problems an N-level atom or molecule absorbs energy from near-resonance monochromatic laser fields, and one asks for the time-dependent populations of the energy levels of the absorber. We find that the equal-Rabi and the harmonic-Rabi N-level systems studied numerically in the preceding paper are associated with Chebyshev and Hermite polynomials. Using the exact solutions we find exact expressions for the time averages of occupation probabilities in terms of sums of squares of polynomials. In addition we find the first approximate analytic expression for the long-period population oscillations observed in numerical solutions; and we show that a steady-state population inversion between the highest and lowest levels may be maintained by multiphoton pumping.

I. INTRODUCTION

Numerical studies of laser-induced atomic or molecular excitation, described in the preceding paper,[1] have shown the new features of many-level systems which are not present in simple two- or three-level atomic or molecular models. These indicate that it is worthwhile to develop a broad framework for the analytic study of multilevel systems interacting with many laser beams for arbitrary level multiplicity. In this paper we give a general, but still very simple, theory of multilevel systems interacting with many intense photon beams which are in (or near) resonance with the transitions occurring between neighboring levels. The simplicity of this theory is due to our extreme point of view: that for sufficiently intense beams only coherent interactions are important and one can neglect all losses due to incoherent relaxation. We also use the global rotating-wave approximation (RWA) and neglect all nonresonant interactions.

Our theory is based on the mathematical equivalence of the following two classes of physical problems: (i) an N-level quantum system interacting with $N-1$ photon beams in RWA, (ii) a chain of N classical harmonic oscillators interacting via nearest-neighbor harmonic forces (the so-called Sturm system).

The basic mathematics encountered in both classes reduces to the study of Jacobi matrices. Such studies were made in the past in connection with Sturm systems,[2] and we will use the results of these studies to draw conclusions relevant for coherent optical interactions of multilevel systems.

The general theory is developed in Sec. II and special cases (including those studied in ESBB, Ref. 1) are described in some detail in Sec. III. Section IV is devoted to studies of the time averages of level populations, which are found to be particularly amenable to our analysis, and Sec. V provides a compact analytic explanation of the basic long period for the "population sloshing" observed in ESBB, Sec. III.

II. N-LEVEL SYSTEM AND JACOBI MATRICES

The semiclassical time-dependent Hamiltonian describing the fully coherent interaction of the N-level quantum system ("atom") with $N-1$ laser beams in RWA can be written in the form

$$H(t)=\sum_{m=1}^{N}E_m b_m^\dagger b_m+\frac{1}{2}\sum_{m=1}^{N-1}\hbar\Omega_m b_{m+1}^\dagger b_m e^{-i\omega_m t}+\text{H.c.} \tag{1}$$

where b_m ($b_m^\dagger$) annihilates (creates) the atom or molecule in the mth unperturbed state $|m\rangle$ of energy E_m, ω_m is the frequency of the mth laser beam, and Ω_m is the Rabi frequency defined in ESBB (4).

The Hamiltonian (1) can be transformed to a time-independent form by the following unitary transformation,

$$U(t)=\exp\left\{-it\left[\sum_{m=1}^{N}\left(\sum_{k=1}^{m-1}\omega_k+D_0\right)b_m^\dagger b_m\right]\right\}, \tag{2}$$

where D_0 is arbitrary. In the representation of unperturbed states, this transformed Hamiltonian can be written as the following $N\times N$ tridiagonal time-independent matrix:

$$\begin{vmatrix} D_0 & \frac{1}{2}\Omega_1 & 0 & \cdots & & \\ \frac{1}{2}\Omega_1^* & D_1 & \frac{1}{2}\Omega_2 & & & \\ 0 & \frac{1}{2}\Omega_2^* & D_2 & & & \\ \cdot & & \cdot & & & \\ \cdot & & & \cdot & & \\ \cdot & & & & \cdot & \\ & & & & D_{N-2} & \frac{1}{2}\Omega_{N-1} \\ & & & & \frac{1}{2}\Omega_{N-1}^* & D_{N-1} \end{vmatrix}, \tag{3}$$

where D_m denotes the accumulated detuning of the mth transition defined in ESBB. The Schrödinger equation (ESBB-1) is based on this form of Hamiltonian.

It is convenient to have all equations dimensionless. We choose to do this by scaling the time by the modulus of the Rabi frequency of the first transition,

$$\tau=|\Omega_1|\,t\,, \tag{4}$$

and by referring all other frequencies and detunings to this Rabi frequency.

By an appropriate choice of the phases of the basis vectors $|m\rangle$, one can transform the possibly complex matrix (3) to the real Jacobi form,[3]

$$\mathcal{J}_N=\begin{vmatrix} \Delta_1 & c_1 & 0 & \cdots & & \\ c_1 & \Delta_2 & c_2 & & & \\ 0 & c_2 & \Delta_3 & & & \\ \cdot & & \cdot & & & \\ \cdot & & & \cdot & & \\ \cdot & & & & \cdot & \\ & & & & \Delta_{N-1} & c_{N-1} \\ & & & & c_{N-1} & \Delta_N \end{vmatrix}, \tag{5}$$

where

$$\Delta_m=D_{m-1}/|\Omega_1| \quad\text{and}\quad c_m=\tfrac{1}{2}|\Omega_m|/|\Omega_1|\,. \tag{6}$$

are real quantities and c_m is positive.

In order to solve the problem of the time evolution of the state vectors, we have to find the eigenvalues and eigenvectors of the matrix $\mathcal{J}_N$. As we have mentioned in Sec. I, the same mathematical problem arises in classical mechanics in the study of coupled harmonic oscillators. Many of the properties of Jacobi matrices found in connection with their role in classical mechanics turn out to be of importance in the study of the optical interactions of multilevel quantum systems as well. We will give a brief review of those properties which will be used in Sec. III.

Let us introduce a set of N polynomials $\mathfrak{D}_m(\lambda)$, defined as the determinants of the $N-1$ principal minors associated with a given Jacobi matrix $\mathcal{J}_N$, along with the "initial" polynomial $\mathfrak{D}_0(\lambda)=1$.

$$\begin{aligned} &\mathfrak{D}_0(\lambda)=1\,,\\ &\mathfrak{D}_1(\lambda)=\det(\Delta_1-\lambda)=\Delta_1-\lambda\,,\\ &\mathfrak{D}_2(\lambda)=\det\begin{pmatrix}\Delta_1-\lambda & c_1\\ c_1 & \Delta_2-\lambda\end{pmatrix}=(\Delta_1-\lambda)(\Delta_2-\lambda)-c_1^2,\\ &\mathfrak{D}_3(\lambda)=\det\begin{pmatrix}\Delta_1-\lambda & c_1 & 0\\ c_1 & \Delta_2-\lambda & c_2\\ 0 & c_2 & \Delta_3-\lambda\end{pmatrix}\\ &\qquad=(\Delta_1-\lambda)(\Delta_2-\lambda)(\Delta_3-\lambda)\\ &\qquad\quad-c_1^2(\Delta_3-\lambda)-c_2^2(\Delta_1-\lambda)\,, \end{aligned} \tag{7}$$

and so on. The polynomials $\mathfrak{D}_m(\lambda)$ obey the following recurrence relations, for $m\geq 1$:

$$\mathfrak{D}_{m+1}(\lambda)=(\Delta_{m+1}-\lambda)\,\mathfrak{D}_m(\lambda)-c_m^2\,\mathfrak{D}_{m-1}(\lambda)\,. \tag{8}$$

Clearly, $\mathfrak{D}_N(\lambda)$ is the characteristic polynomial of the original Jacobi matrix $\mathcal{J}_N$. Therefore the roots $\lambda_1,\ldots,\lambda_N$ of $\mathfrak{D}_N(\lambda)$ are the eigenvalues of $\mathcal{J}_N$,

$$\mathfrak{D}_N(\lambda_k)=0\,. \tag{9}$$

If all coefficients c_m are different from zero, all these roots are different.

The eigenvectors of $\mathcal{J}_N$ have a simple form when expressed in terms of the polynomials $\mathfrak{D}_m(\lambda)$. We will denote the normalized eigenvectors by $|\lambda_k\rangle$. The scalar products $\langle m|\lambda_k\rangle$ are the transformation coefficients connecting the unperturbed basis states $|m\rangle$ with the new basis states $|\lambda_k\rangle$. These states $|\lambda_k\rangle$ are the eigenstates of the total Hamiltonian in the global rotating-wave approximation (that is, the dressed-atom states). For future convenience in writing we define

$$\begin{aligned} v_{m+1}(\lambda_k)&=\langle m+1|\lambda_k\rangle\\ &=(-1)^m A(\lambda_k)(c_0c_1\ldots c_m)^{-1}\mathfrak{D}_m(\lambda_k)\,, \end{aligned} \tag{10}$$

where $c_0\equiv 1$, and

$$A(\lambda_k)=\left(\sum_{m=0}^{N-1}(c_0c_1\ldots c_m)^{-2}[\mathfrak{D}_m(\lambda_k)]^2\right)^{-1/2}. \tag{11}$$

An arbitrary vector $|\Psi(\tau)\rangle$ evolves in time according to the formula

$$|\Psi(\tau)\rangle=\sum_{k=1}^{N}e^{-i\lambda_k\tau}|\lambda_k\rangle\langle\lambda_k|\Psi(0)\rangle\,. \tag{12}$$

The transition amplitude $T_{mn}(\tau)$ to find the atom in its mth unperturbed state $|m\rangle$ at time τ, if it was at $\tau = 0$ in its nth state $|n\rangle$ is

$$T_{m,n}(\tau) = \sum_{k=1}^{N} e^{-i\lambda_k \tau} \langle m|\lambda_k\rangle \langle \lambda_k|n\rangle = \sum_{k=1}^{N} e^{-i\lambda_k \tau} v_m(\lambda_k) v_n^*(\lambda_k) . \quad (13)$$

The transition probability is therefore given by

$$P_{mn}(\tau) = \sum_{k,l=1}^{N} e^{-i(\lambda_k - \lambda_l)\tau} v_m^*(\lambda_l) v_m(\lambda_k) v_n(\lambda_l) v_n^*(\lambda_k) . \quad (14)$$

Since the eigenvalues λ_k are in general not commensurate, changes of populations with time will be periodic only for very special choices of the Hamiltonian matrix.

III. SPECIAL SOLUTIONS FOR N-LEVEL SYSTEMS

Since the relation (8) connecting the polynomials $\mathfrak{D}_m(\lambda)$ have the form of recurrence relations for orthogonal polynomials, we would expect to obtain particularly simple ("exactly solvable") results whenever the recurrence (8) coincides with that of a known classical polynomial. We found the cases of Chebyshev, Hermite, Legendre, and Laguerre polynomials especially interesting.

A. Chebyshev polynomials

This case corresponds to an interaction with no detuning ($\Delta_m = 0$) and with equal Rabi frequencies ($c_m = \frac{1}{2}$). This is one of the problems extensively studied numerically in ESBB. The solution of its mechanical counterpart goes back to Euler, d'Alembert, and Bernoulli.[4]

The components $\mathfrak{D}_m(\lambda_k)$ of the (unnormalized) eigenvectors are proportional to the Chebyshev polynomials of the second kind,

$$\mathfrak{D}_m(\lambda) = (-\tfrac{1}{2})^m U_m(\lambda) . \quad (15)$$

These polynomials can be expressed in terms of trigonometric functions in the following way,[5]

$$U_m(\cos\Theta) = \sin[(m+1)\Theta]/\sin\Theta . \quad (16)$$

It follows from this formula and Eq. (9) that the eigenvalues λ_k are

$$\lambda_k = \cos[k\pi/(N+1)], \quad k = 1, 2, \ldots, N , \quad (17)$$

and that the components of the normalized eigenvectors can be written

$$v_m(\lambda_k) = \left(\frac{2}{N+1}\right)^{1/2} \sin\frac{mk\pi}{N+1}, \quad m = 1, 2, \ldots, N . \quad (18)$$

The amplitude $T_{m1}(\tau)$ which determines the population in level m evolving from the ground level $n = 1$ is given by

$$T_{m1}(\tau) = \frac{2}{N+1} \sum_{k=1}^{N} \exp\left(-i\tau \cos\frac{k\pi}{N+1}\right) \times \sin\frac{km\pi}{N+1} \sin\frac{k\pi}{N+1} . \quad (19)$$

In the special case of the five-level system studied numerically in ESBB the eigenvalues are $\frac{1}{2}\sqrt{3}$, $\frac{1}{2}, 0, -\frac{1}{2}, -\frac{1}{2}\sqrt{3}$ and the transition amplitude to the middle level only is a periodic function of time,

$$T_{31}(\tau) = \tfrac{1}{3}[-1 + \cos(\tfrac{1}{2}\tau\sqrt{3})] = -\tfrac{2}{3}\sin^2(\tau\sqrt{3}/4) . \quad (20)$$

All remaining transition amplitudes $T_{m1}(\tau)$, $m \neq 3$, will contain both of the incommensurate frequencies $\frac{1}{2}$ and $\frac{1}{2}\sqrt{3}$ and therefore will not change periodically in time.

B. Hermite polynomials

Hermite polynomials emerge if there is no detuning and the sequence of Rabi frequencies is $\sqrt{1}, \sqrt{2}, \sqrt{3}, \ldots$ ($c_m = \frac{1}{2}\sqrt{m}$). This situation arises physically if the N-level system comprises the N lowest levels of a harmonic oscillator. This case has been studied by Feldman and Elliott[6] and in ESBB. The recurrence relation in this case is

$$\mathfrak{D}_{m+1}(\lambda) = -\lambda\mathfrak{D}_m(\lambda) - \tfrac{1}{4} m \mathfrak{D}_{m-1}(\lambda) , \quad (21)$$

and it leads to the following result:

$$\mathfrak{D}_m(\lambda) = (-1)^m 2^{-3m/2} H_m(\sqrt{2}\lambda) . \quad (22)$$

From the Christoffel-Darboux formula for the Hermite polynomials[5] one obtains the following formula for the normalized eigenvectors,

$$v_{m+1}(\lambda_k) = |N H_{N-1}(\sqrt{2}\lambda_k)|^{-1} \times (N!\, 2^{N-m-1}/m!)^{1/2} H_m(\sqrt{2}\lambda_k) . \quad (23)$$

The eigenvalues λ_k are $1/\sqrt{2}$ times the roots of the Nth Hermite polynomial.

C. Legendre polynomials

This case also corresponds to no detuning, and the coefficients c_m must be chosen in the form

$$c_m = (4 - 1/m^2)^{-1/2} . \quad (24)$$

That is, the dimensionless Rabi frequencies quickly approach unity from above as m gets large. The normalized eigenvectors are

$$v_{m+1}(\lambda_k) = |N P_{N-1}(\lambda_k)|^{-1} (2m+1)^{1/2} (1-\lambda_k^2)^{1/2} P_m(\lambda_k) , \quad (25)$$

and the eigenvalues are roots of P_N.

D. Laguerre polynomials

This is the simplest case with detuning. The coefficients c_m and Δ_m in this case are

$$c_m = m\,, \quad \Delta_m = 2m - 1\,. \tag{26}$$

Comparing Eq. (26) with Eqs. (6) and (ESBB-3), we see that all detuning frequencies are the same and equal to $2|\Omega_1|$.

The components of the normalized eigenvectors in this case are

$$v_{m+1}(\lambda_k) = |NL_{N-1}(\lambda_k)|^{-1}\lambda_k^{-1/2}L_m(\lambda_k)\,, \tag{27}$$

and the eigenvalues λ_k are the roots of L_N.

The relations described here, and similar ones associated with other polynomials, indicate one answer to the question posed in ESBB. A general framework has been found within which whole classes of N-level problems can be discussed. In addition, we will show in the next section that the polynomial method easily provides some quantitative results about long-time behavior not readily obtained by *ab initio* numerical integration.

IV. TIME-AVERAGED LEVEL POPULATIONS

The time dependence of the level occupation probabilities, described by Eq. (14) and also revealed by the numerical studies of the preceding paper, becomes very complicated for large N. Obviously not every detail of this time dependence has great physical significance. We will certainly simplify the description greatly by considering averaged characteristics of the atomic system. The simplest approach consists in evaluating time averages p_{mn} of transition probabilities over infinite time intervals:

$$p_{mn} = \lim_{T\to\infty}\frac{1}{T}\int_0^T dt\, P_{mn}(t)\,. \tag{28}$$

These probabilities measure the relative average time that the atom spends in its mth unperturbed state if it was originally in its nth unperturbed state.

All oscillating terms drop out as a result of time averaging, because all eigenvalues λ_k are different, and the final formula for p_{mn} is

$$p_{mn} = \sum_{k=1}^{N} |v_m(\lambda_k)|^2\,|v_n(\lambda_k)|^2\,. \tag{29}$$

This expression can be evaluated in a closed form for the equal-Rabi case,

$$p_{mn} = \left(\frac{2}{N+1}\right)^2\sum_{k=1}^{N}\left(\sin\frac{nk}{N+1}\right)^2\left(\sin\frac{mk}{N+1}\right)^2$$

$$= \frac{1}{N+1}\left(1+\tfrac{1}{2}\delta_{mn}+\tfrac{1}{2}\delta_{m,N+1-n}\right). \tag{30}$$

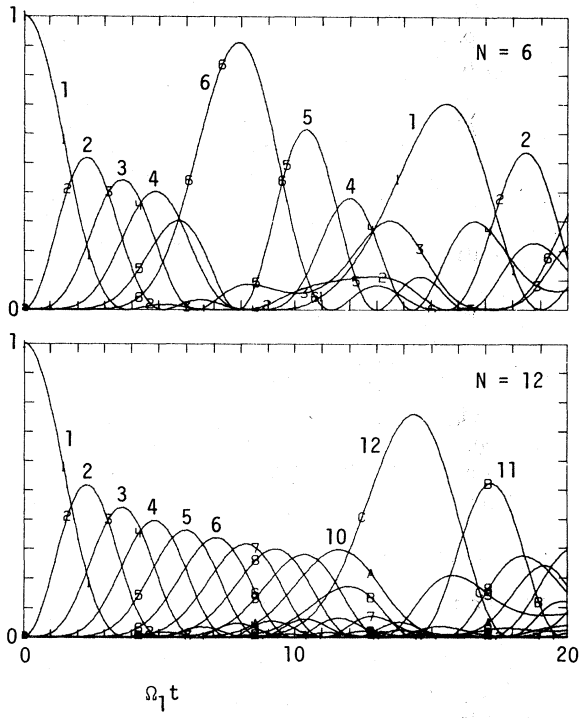

FIG. 1. Populations of the levels of 6-level and 12-level atoms in the equal-Rabi case.

It follows from this formula that, on the average, all levels with the exception of the initially excited level (level n) and its mirror image (level $N+1-n$) are equally populated. The relative probability of the exceptional levels is $\frac{3}{2}$ times larger. In the special case when $n = N+1-n$, the population of the initial level is twice the population of the other levels. This average behavior is a consequence of the kind of temporal behavior shown in Fig. 1 and in ESBB, Fig. 2, for the equal-Rabi case.

In the harmonic-Rabi case, the formula for the averaged probabilities reads

$$p_{m+1,n+1} = (N!)^2 N^{-4} 4^{N-1}\sum_{k=1}^{N}[H_{N-1}(\sqrt{2}\lambda_k)]^{-4}\,2^{-m-n}$$
$$\times (m!)^{-1}(n!)^{-1}[H_m(\sqrt{2}\lambda_k)]^2$$
$$\times [H_n(\sqrt{2}\lambda_k)]^2\,. \tag{31}$$

In Fig. 2 we show the time-averaged probabilities for the levels of an equal-Rabi system and of a harmonic-Rabi system having 2, 3, 4, 5, 7, and 15 levels. These graphs make it clear that the temporal behavior, shown for only a few population cycles in Fig. 1, is a fairly general property of the interaction, not associated solely with the first population surge. That is, the two ends of the excitation ladder are preferred locations for the probability over the long term as well as initially.

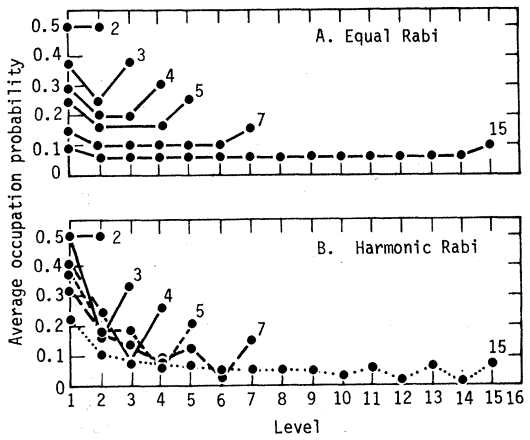

FIG. 2. Time-averaged level populations of the levels of 2, 3, 4, 5, 7, and 15-level systems, in both the equal-Rabi and the harmonic-Rabi cases.

(When the Rabi frequencies are successively smaller in a non-equal-Rabi system the intermediate levels are dominant in the long term.)

V. LONG-PERIOD POPULATION OSCILLATION

One of the striking features observed by ESBB, and evident here in Fig. 1, was the relatively slow oscillation of population from one end of the excitation ladder to the other and back again. The period of this "sloshing" oscillation is significantly greater than any Rabi period associated directly with the system-laser interaction. Such slow, and fairly stable, near-recurrences of the initial configuration indicate again the persistent internal coherence of the dressed states of the system.

We can show the origin of the long-period slosh-ing in the equal-Rabi case, and can evaluate the period itself approximately. For an equal-Rabi system the expression for the transition amplitude from the lowest to the highest level is:

$$T_{N1}(\tau) = \frac{2}{N+1} \sum_{k=1}^{N} (-1)^{k+1} \exp(-i\tau \cos\varphi_k) \sin^2\varphi_k \ , \tag{32}$$

where

$$\varphi_k = k\pi/(N+1) \ . \tag{33}$$

The weight function $\sin^2\varphi_k$ enhances contributions from the slowly varying terms, those with small $\cos\varphi_k$, i.e., φ_k close to $\pi/2$. These small eigenvalues are nearly equidistant and their spacing is close to $\pi/(N+1)$ because

$$\cos\varphi_{(N+1)/2-l} \simeq \frac{\pi}{2} \frac{N+1-2l}{N+1} \quad \text{for small } l \ . \tag{34}$$

For that reason the amplitude T_{N1} will exhibit a quasiperiodic behavior with half-period equal to $N+1$. One can see this explicitly by evaluating the sum in Eq. (32) in a closed form in the approximation (34). Using standard relations for trigonometric functions, we obtain in this approximation:

$$T_{N1}(\tau) = \tfrac{1}{2} i \left[e^{-i\pi\tau/2} + (-1)^N e^{i\pi\tau/2} \right] \tan\frac{\pi\tau}{2(N+1)} \times \frac{\cos[2\pi/(N+1)] - 1}{\cos[2\pi/(N+1)] + \cos[\pi\tau/(N+1)]} \ . \tag{35}$$

This formula describes fairly well the many cycles of slow pulsation in the populations of the highest and lowest levels evident in the graphs in Fig. 3. For small τ (relative to N) the amplitude T_{N1} stays close to zero and it reaches its first maximum, equal to one in our crude approximation, at $\tau = N+1$. As time goes on, departures from the equal-spacing formula (34) increase, of course, and aperiodic terms will eventually destroy the population sloshing.

The approximate expression derived here for the sloshing period shows a linear relation between the period and the highest level number. This is consistent with Fig. 7 in ESBB, where it is seen that the time required for the population in level N to reach its first maximum is approximately linear in

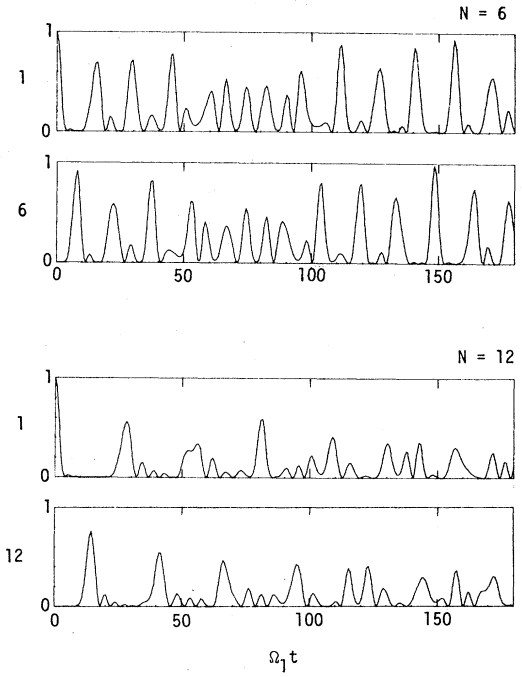

FIG. 3. Populations of the lowest and highest levels of 6-level and 12-level atoms in the equal-Rabi case, showing the long-period population sloshing from the bottom to the top of the excitation ladder and back again.

N. It is interesting to compare this finding with the result to be expected of an infinite (i.e., untruncated) equal-Rabi atom or molecule. This can be done indirectly.

The probability of occupation of level m at the time $\tau = \Omega_1 t$ is given for the untruncated equal-Rabi case in (ESBB-9):

$$P_{m,1}(t) = [J_{m-1}(\tau)]^2 , \tag{36}$$

where, as in (ESBB-9), m is in this case infinite in range: $-\infty \leq m \leq \infty$. We now calculate the average location of probability at time τ, actually the average departure from the initial value $m = 1$:

$$\langle |m-1| \rangle = \sum_{m=-\infty}^{\infty} |m-1| P_{m,1}(t) \tag{37}$$

we find[7]

$$\langle |m-1| \rangle = \tfrac{1}{2}\tau \{ \tfrac{1}{4}\tau [J_0^2(\tau) + J_1^2(\tau)] - J_0(\tau) J_1(\tau) \} . \tag{38}$$

In Fig. 4 we plot $\langle |m-1| \rangle$ vs τ and find a relation that is very nearly exactly linear. That is, the average of the population flow upward in an untruncated equal-Rabi atom or molecule obeys a law very much like that obeyed by a truncated one. Detailed comparisons of both equal-Rabi and harmonic-Rabi N-level systems with their untruncated counterparts indicate that the latter can serve as useful simple approximations to the former in some circumstances.[8]

VI. TIME-AVERAGED INVERSION

The possibility of inversion in the level populations is always interesting. In ESBB and in Secs. IV and V here we have described the relatively large population of the last level, even in the long-time average.

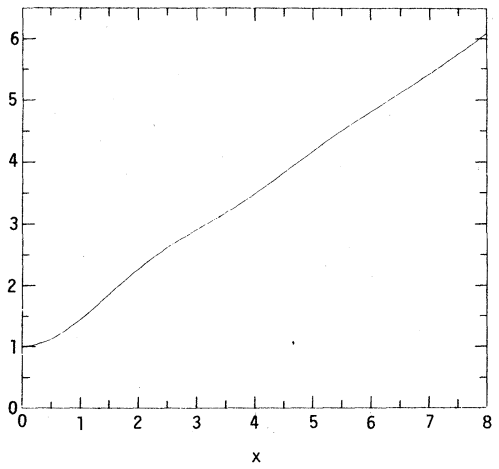

FIG. 4. Average number of transitions made away from the initial ground state as a function of time, for an infinite-level equal-Rabi atom.

However, there is no time-averaged population inversion between the ground level and the highest level in the equal-Rabi case or in the harmonic-Rabi case. The question arises: Can one obtain such a steady-state population inversion by a proper choice of Rabi frequencies and detunings? That is, could one promote very high harmonics of the pumping laser to appear in fluorescence from high levels? We do not know the answer to this question in the general case since it would require a solution of highly complicated algebraic inequalities. We can, however, answer this question completely for three-level and four-level atoms with no detunings, since we can evaluate the averaged probabilities p_{mn} in closed form.

Here we give a list of eigenvalues and time-averaged transition probabilities for three- and four-level systems with no detuning and *arbitrary* Rabi frequencies ($c_m = \frac{1}{2}|\Omega_m| / |\Omega_1|$). In the three-level case we have

$$\lambda_1 = (c_1^2 + c_2^2)^{1/2}, \quad \lambda_2 = 0, \quad \lambda_3 = -(c_1^2 + c_2^2)^{1/2}, \tag{39}$$

$$p_{11} = \tfrac{1}{2}(c_1^4 + 2c_2^4)(c_1^2 + c_2^2)^{-2}, \tag{40}$$

$$p_{21} = \tfrac{1}{2} c_1^2 (c_1^2 + c_2^2)^{-1}, \tag{41}$$

$$p_{31} = \tfrac{1}{2}\, 3 c_1^2 c_2^2 (c_1^2 + c_2^2)^{-2}, \tag{42}$$

and, in the four-level case,

$$\lambda_{1,2} = \pm 2^{-1/2} (c_1^2 + c_2^2 + c_3^2 + r^{1/2})^{1/2}, \tag{43}$$

$$\lambda_{3,4} = \pm 2^{-1/2} (c_1^2 + c_2^2 + c_3^2 - r^{1/2})^{1/2}, \tag{44}$$

$$p_{11} = (c_1^4 + c_2^4 + c_3^4 + 2c_2^2 c_3^2 - 2c_1^2 c_3^2)(2r)^{-1}, \tag{45}$$

$$p_{21} = (c_1^4 + c_3^4 + c_1^2 c_2^2 + c_2^2 c_3^2 - 2c_1^2 c_3^2)(2r)^{-1}, \tag{46}$$

$$p_{31} = c_1^2 c_2^2 r^{-1}, \tag{47}$$

$$p_{41} = c_2^2 (c_1^2 + c_2^2 + c_3^2)(2r)^{-1}, \tag{48}$$

where

$$r = (c_1^2 + c_2^2 + c_3^2)^2 - 4c_1^2 c_3^2 . \tag{49}$$

The ratio of the highest to the lowest level population is, for the three-level atom,

$$\frac{p_{31}}{p_{11}} = \frac{3c_1^2 c_2^2}{c_1^4 + 2c_2^4} , \tag{50}$$

and for the four-level atom,

$$\frac{p_{41}}{p_{11}} = \frac{c_2^2 (c_1^2 + c_2^2 + c_3^2)}{c_1^4 + c_2^4 + c_3^4 + 2c_2^2 c_3^2 - 2c_1^2 c_3^2} . \tag{51}$$

The maximal values of these ratios are 1.05 for the three-level atom and 1.2 for the four-level atom.[9]

VII. SUMMARY

In this paper we have developed a general framework for the analysis of multilevel quantum systems undergoing near-resonance excitation by one or more monochromatic radiation fields. We have been able to do this compactly by recognizing the mathematical equivalence of the classical problem of the Sturm system. Transition amplitudes, and particularly time-averaged transition probabilities, have simple expressions in terms of the eigenvalues and eigenfunctions of certain Jacobi matrices. We have shown that in several cases, explored extensively numerically, these eigenfunctions are well-known classical orthogonal polynomials. We have also been able to explain qualitatively the long-period population "sloshing" observed in the preceding paper (ESBB), and in the equal-Rabi case have given an approximate analytic formula for the value of the period. Finally, we have shown that if the Rabi frequencies are properly adjusted in three-level and four-level systems, a steady-state population inversion may be induced between the highest and lowest levels by the multilaser pumping of the systems.

ACKNOWLEDGMENTS

The first two authors would like to acknowledge the hospitality extended to them at the University of Rochester.

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