Coherent dynamics of N-level atoms and molecules. III. An analytically soluble periodic case

Richard J. Cook and Bruce W. Shore
University of California, Lawrence Livermore Laboratory, P. O. Box 808, Livermore, California 94550
(Received 13 March 1979)

By employing an analogy with a spin-J system in a constant magnetic field the authors obtain a simple soluble model for stepwise laser excitation of an N-level system ($N = 2J + 1$), including analytic treatment of Doppler detuning and of ionization loss. The solutions are periodic and hence permit complete population inversion. A simple graphical realization of the population dynamics, a generalization of the Feynman, Vernon, and Hellwarth vector model of the two-level Bloch equation, is then described.

INTRODUCTION

The dynamics of incoherent radiative atomic excitation, known since the days of Einstein,[1-4] differs qualitatively from the behavior of systems excited coherently: retention of phase memory during coherent excitation leads to oscillatory or fluctuating excited-state populations, whereas incoherently excited populations typically tend monotonically toward equilibrium values.[5]

The increasing experimental study of single- and multiple-laser excitation, in atoms and in molecules,[6,7] brings growing interest in simple analytically soluble theoretical models of the excitation dynamics.[8-10] Over the last few years many authors have explored aspects of one such model, the N-level atom (or molecule), which idealizes a linkage of excitations driven simultaneously be monochromatic light tuned to (or near) the Bohr resonance frequencies. (Some authors have examined general formalism,[11-19] others have examined numerical and analytical results applied to molecular multiphoton excitation,[20-28] while others have described simple analytically soluble cases.[29-35]) In elementary form the model expresses the population $P_n(t)$ of level n at time t (for $1 \leq n \leq N$) as the absolute square of a complex-valued probability amplitude $C_n(t)$ obtained as the solution to the rotating-wave-approximation (RWA) time-dependent Schrödinger equation

$$i\frac{d}{dt}C_n(t) = \sum_m W_{nm}C_m(t) . \tag{1}$$

For the simple cases considered in the present paper,[29-35] stepwise excitation of nondegenerate levels, the RWA Hamiltonian W is tridiagonal. Diagonal elements W_{nn} have as their real parts the cumulative detuning Δ_n of $n-1$ successive lasers away from the corresponding sum of $n-1$ Bohr frequencies, and have as their negative imaginary parts half the probability loss rate γ_n from level n (e.g., ionization loss or spontaneous emission to a level not included in the N-level sequence):

$$W_{nn} = \Delta_n - \tfrac{1}{2}i\gamma_n . \tag{2}$$

The nonzero off-diagonal elements of W, those linking adjacent levels, are proportional to the dipole transition moment d_n linking levels in n and $n+1$ and to the (possibly complex-valued) electric field amplitude $\mathcal{E}_n$ whose carrier frequency matches the Bohr frequency for the $n \rightarrow n+1$ transition:

$$W_{n,n+1} = -(1/\hbar)d_n\mathcal{E}_n^* = W_{n+1,n}^* \equiv \tfrac{1}{2}\Omega_n . \tag{3}$$

Following now common usage, we refer to the parameter Ω_n as the Rabi frequency for step n. Thus the N-level atom comprises a sequence of $N-1$ values for the parameters Ω_n and of N values for the parameters Δ_n, γ_n. In principle each of these quantities may be fixed arbitrarily: the Rabi frequency by adjusting laser intensity, the detuning by adjusting laser frequency, and the loss, if any, by imposing additional ionizing lasers. This N-level model, though highly idealized as a model for laser excitation, provides useful insights into the details of coherent excitation and into the relationship between the physically adjustable parameters and population variations. Equally important, simple analytic solutions can provide a valuable check upon purely numerical methods of solution.

A traditional method for solving systems of linear first-order ordinary differential equations with constant coefficients, Eq. (1), proceeds through the construction of eigenvectors of the coefficient matrix W:

$$\sum_m (W_{nm} - \lambda_k\delta_{nm})\langle m|\lambda_k\rangle = 0 . \tag{4}$$

Using these, one readily constructs the time evolution matrix $\exp(-iWt)$ and so obtains the expression for probability amplitude $C_n(t)$ as

$$C_n(t)=\sum_k\sum_m\langle n|\lambda_k\rangle\exp(-i\lambda_k t)\langle\lambda_k|m\rangle C_m(0)\,. \quad (5)$$

This formula expresses the solution $C_n(t)$ as a discrete Fourier series, with frequencies equal to the eigenvalues λ_k of the matrix W. One readily sees that, in general, populations vary periodically in time only if the eigenvalues are commensurable (exceptional cases of periodicity occur when eigenvector components vanish). Thus, although the two- and three-level resonant ($\Delta_n=0$) lossless ($\gamma_n=0$) atoms are strictly periodic, the solutions for $N>3$ are not generally strictly periodic.[32,33] (Solutions will, of course, come arbitrarily close to repetition over sufficiently long time.) It is natural to ask if general cases can be prescribed which are periodic for $N>3$. The present paper describes such a case.

The constructive algorithm presented in Eqs. (4) and (5) poses no difficulty for a digital computer, and one can in practice readily evaluate numerical solutions for arbitrary values of the model parameters and for N as large as several tens. Nevertheless, it is useful to know particular parameter choices which admit tractable analytic solutions, i.e., those for which the eigenvectors are expressible in terms of well-known special functions. Table I lists the previously reported[29,33-35] analytically soluble cases and names the functions which provide the eigenvectors in these cases. For completeness we include, as a final line, the present-model solutions. In none of the previously studied cases are the solutions generally strictly periodic for $N>3$. (An exception occurs for level 3 of the 5-level atom having equal Rabi frequencies.[32,33]) Furthermore, none of the previously studied cases include probability loss. The following sections describe a simple analytically soluble case which is strictly periodic and allows both detuning and probability loss of a special form.

I. SPIN MODEL

Our model rests upon mathematical analogy with a spin-J system in a constant magnetic field. We identify the number of atom levels N with the spin degeneracy $2J+1$, and we identify the nth level with a specific magnetic sublevel labeled by eigenvalue M of J_z:

$$N=2J+1,\quad J=\tfrac{1}{2}(N-1),$$
$$n=M+J+1,\quad M=n-\tfrac{1}{2}(N+1).$$

We thereby create a one-to-one correspondence between basis states $|Nn\rangle$ and those of a pseudo-angular-momentum representation $|JM\rangle$:

$$|Nn\rangle=|JM\rangle\,. \quad (6)$$

It is now a simple matter to evaluate matrix elements of the operator

$$\hbar W=aJ_z+bJ_x+cJ_y+d \quad (7)$$

first in the JM basis and then, by change of notation, in the Nn basis. The matrix W is tridiagonal, with diagonal elements

$$W_{nn}=Ma+d$$
$$=na+[d-\tfrac{1}{2}a(N-1)] \quad (8)$$

and off-diagonal elements

$$W_{n+1,n}=W^*_{n,n+1}=\tfrac{1}{2}(b+ic)\sqrt{J(J+1)-M(M+1)}$$
$$=\tfrac{1}{2}(b+ic)\sqrt{n(N-n)}\,. \quad (9)$$

Thus this model represents the sequence of Rabi frequencies

$$\Omega_n=\Omega_0\sqrt{n(N-n)}\,, \quad (10)$$

where Ω_0 is an arbitrary complex-valued scale factor; the detunings and loss rates vary linearly with excitation n:

$$\Delta_n=n\Delta_0+D_0,\quad \gamma_n=n\gamma_0\,, \quad (11)$$

where Δ_0 and D_0 are arbitrary real numbers and γ_0 is a non-negative real number. [$D_0=d-\frac{1}{2}a(N-1)$]. It should be emphasized that the formal analogy with a spin-J system places no restriction on the energy levels E_n ($n=1,\ldots,N$) of the N-level atom. The energy levels and associated Bohr frequencies do not appear explicitly in the above formalism, and hence are entirely arbitrary.

To construct eigenvectors of W we need only recognize that W, acting upon basis states $|Nn\rangle$ or $|JM\rangle$, behaves as an angular momentum vector having Cartesian components a, b, c. We introduce a coordinate system oriented along this vector, i.e., rotated by Euler angles (α,β,γ). The length of W is $(a^2+b^2+c^2)^{1/2}$, so that in the new (primed) coordinate system we have the simple expression

$$W'=(a^2+b^2+c^2)^{1/2}J'_z+d\,. \quad (12)$$

The matrix W' is diagonal in this aligned coordinate system. The eigenvalues are simply

TABLE I. Analytic N-level solutions.

Ω_n	Δ_n	γ_n	Eigenvectors
Ω_0	0	0	Chebyshev
$\Omega_0\sqrt{n}$	0	0	Hermite
$\Omega_0(4-n^{-2})^{-1/2}$	0	0	Legendre
$\Omega_0 n$	$2n-1$	0	Laguerre
$\Omega_0[n(N-n)]^{1/2}$	$\Delta_0 n$	$\gamma_0 n$	Jacobi

$$\lambda_n = M(a^2+b^2+c^2)^{1/2}+d$$
$$\equiv n\lambda_0+\Lambda\,. \tag{13}$$

That is, the eigenvalues are evenly spaced at integral values of the basic frequency

$$\lambda_0=(a^2+b^2+c^2)^{1/2}$$
$$=[|\Omega_0|^2+(\Delta_0-\tfrac{1}{2}i\gamma_0)^2]^{1/2}\,. \tag{14}$$

The additive constants d and Λ are arbitrary: they express the choice for zero point of energy levels. The simplest choice is $\Lambda=0$, so that $\lambda_n=n\lambda_0$.

Recognizing that a coordinate rotation diagonalizes W, we can immediately express the eigenvectors $|\lambda_n\rangle$ in terms of rotation matrices[36]:

$$|\lambda_n\rangle=\sum_{n'}|Nn'\rangle D^J_{M'M}(\alpha,\beta,0)\,, \tag{15}$$

where

$$J\equiv\tfrac{1}{2}(N-1),\quad M'\equiv n'-\tfrac{1}{2}(N+1),\quad M\equiv n-\tfrac{1}{2}(N+1)\,.$$

The Euler angles specifying the coordinate rotation are

$$\alpha=\arctan(c/b)$$
$$=\arctan[\mathrm{Im}(\Omega_0)/\mathrm{Re}(\Omega_0)]\,, \tag{16}$$
$$\beta=\arctan[(c^2+b^2)^{1/2}/a]$$
$$=\arctan[|\Omega_0|/(\Delta_0-\tfrac{1}{2}i\gamma_0)]\,. \tag{17}$$

In turn we can write the probability amplitude $C_n(t)$ as

$$C_n(t)=\sum_{n'n''}D^J_{MM'}(\alpha,\beta,0)\exp(-iM'\lambda_0 t-idt)$$
$$\times D^J_{M'M''}(\alpha,\beta,0)^*C_{n''}(0)\,, \tag{18}$$

where $n'\equiv M'+J+1$, $n''\equiv M''+J+1$. All amplitudes here include a common time factor $\exp(-idt)$ which originates with our (arbitrary) choice of energy-level zero point; we can incorporate this phase into $C_n(t)$. The remaining factor $\exp(-iM'\lambda_0 t)$ represents a rotation by angle $\lambda_0 t$ about the figure axis. If we now assume all population resides initially in level n_0, so that $C_{n'}(0)=\delta_{n'n_0}$, the preceding expression becomes

$$C_n(t)=\sum_{n'}D^J_{MM'}(\alpha,\beta,\lambda_0 t)D^J_{M'M_0}(\alpha,\beta,0)^*\,. \tag{19}$$

Thus we have the solution expressed in terms of the rotation matrices; these in turn can be written[36] in terms of Jacobi polynomials of argument $\cos\beta$ together with powers of $\cos\frac{1}{2}(\beta)$ and $\sin\frac{1}{2}(\beta)$:

$$D^{(J)}_{M'M}(\alpha,\beta,0)=\exp(-iM'\alpha)\left(\frac{(J+M')!(J-M')!}{(J+M)!(J-M)!}\right)^{1/2}$$
$$\times(\cos\tfrac{1}{2}\beta)^{M'+M}(\sin\tfrac{1}{2}\beta)^{M'-M}$$
$$\times p^{(M'-M,M'+M)}_{J-M'}(\cos\beta)\,. \tag{20}$$

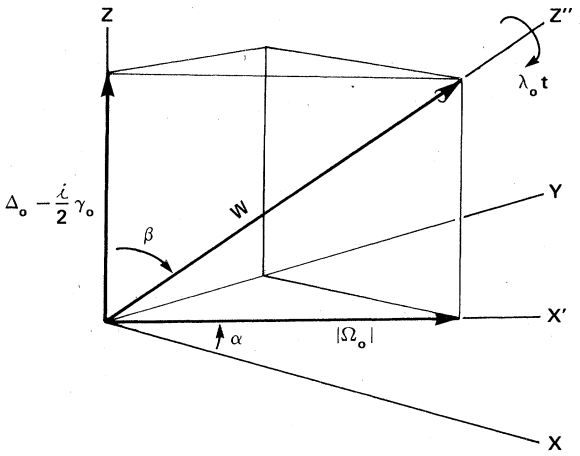

FIG. 1. Representation of the Hamiltonian W as an angular momentum vector: the component in the X-Y plane is the Rabi frequency Ω_0: the Z component is $\Delta_0-\frac{1}{2}\gamma_0$. The Euler angles are α, β, and $\lambda_0 t$.

Figure 1 portrays the relationship between the Hamiltonian W, regarded as an abstract vector, and the Euler angles α, β, and $\gamma=\lambda_0 t$: the magnitude of the projection of W onto the X-Y plane is the Rabi frequency Ω_0, where $\mathrm{Re}(\Omega_0)$ is the X component, $\mathrm{Im}(\Omega_0)$ the Y component; the detuning Δ_0 and loss γ_0 are responsible for the Z component of W. The figure axis Z'' about which steady turning occurs makes an angle β with the vertical.

II. RESONANT EXCITATION

Consider the special case of resonant lossless excitation, $\Delta_0=\gamma_0=0$. Because the eigenvalues are evenly spaced the probabilities are periodic in time with frequency Ω_0. That is, the Rabi frequency serves in this N-level atom, as it does in the two-level atom, both as a measure of interaction strength and of population oscillation frequency. Note that if Ω_0 be fixed then the period is independent of the number of levels N: population flows from level 1 to level N in time $\tau=\pi/\Omega_0$. Figure 2 illustrates this population flow.

Unlike other analytically soluble cases of the N-level atom, the present choice of Rabi frequencies $\Omega_n=\Omega_0\sqrt{n(N-n)}$ leads to *complete population inversion*: if all the population resides in level 1 at time $\tau=0$, then at time $\tau=\pi/\Omega_0$ the population is entirely in level N. The combination of periodicity and complete inversion leads to an N-level "area theorem": let all $N-1$ Rabi frequencies share a common time dependence,

$$\Omega_n=\Omega_0\sqrt{n(N-n)}f(t)\,.$$

Then by defining a new timelike parameter

$$d\tau=dtf(t)\,,$$

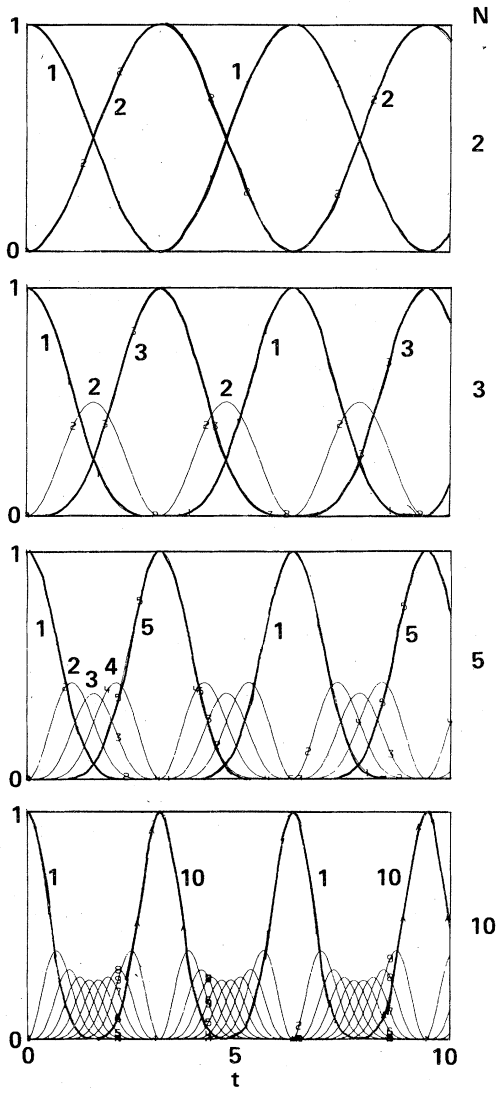

FIG. 2. Populations $P_n(t)$ for N-level spin-model system, for $N=2,3,5$, and 10.

we can recast the RWA Schrödinger equation once again into the form

$$i\frac{d}{d\tau}C_n(t)=\sum_m W_{nm}C_m(t)\,,$$

the solutions of which appear in the preceding section. We thereby deduce that complete N-level inversion occurs for times T such that the area under $f(t)$ is an odd-integer multiple of π:

$$\int_0^T dt\, f(t)=(2K+1)\pi\,.$$

III. N-LEVEL VECTOR MODEL

We have presented in Fig. 1 the representation of the Hamiltonian W as a vector in an abstract three-dimensional space. As a simple generalization of the two-level vector model of Feynman *et al.*[37] we can introduce a second three-dimensional space within which we can represent the population amplitudes of the N-level atom as a vector C. Within this second space we introduce a quantized angular momentum vector of length $[J(J+1)]^{1/2}=\frac{1}{2}(N^2-1)^{1/2}$.

Initially, at time $t=0$, the population resides entirely in some level n. The angular momentum then has a well-defined Z component $M=n-J-1=n-\frac{1}{2}(N+L)$. Because X and Y components of this angular momentum are uncertain, we deal with a *cone* oriented along the Z axis, with apex at the origin, of height M and of base radius

$$[J(J+1)-M^2]^{1/2}=[\tfrac{1}{2}(N-1)(2n-1)-(n-1)^2]^{1/2}\,.$$

That is, the cone is most narrow, having base radius $\sqrt{J}=\sqrt{\frac{1}{2}(N-1)}$, when population resides entirely in either level 1 or level N; when population is entirely in the middle level $n=\frac{1}{2}(N+1)$ of an odd-N system the cone degenerates into a flat disc of radius $\frac{1}{2}(N^2-1)^{1/2}$. Figure 3 illustrates the geometry of the initial-value population cones.

As time progresses the population changes, expressed analytically through the rotation matrix $D^J_{M',M}(\alpha,\beta,\lambda_0 t)$, correspond to a steady turning at rate λ_0 about a figure axis Z'' whose direction in the population space is just the direction fixed by the vector W. When detuning and loss are absent, so that $\beta=90°$, the figure axis is in the X-Y plane (it is the X axis if Ω_0 is real). Under such resonant lossless conditions the population cone precesses about the X axis. After a half period (or π pulse), at time $t=\pi/\lambda_0$, the cone points opposite the initial direction, corresponding to com-

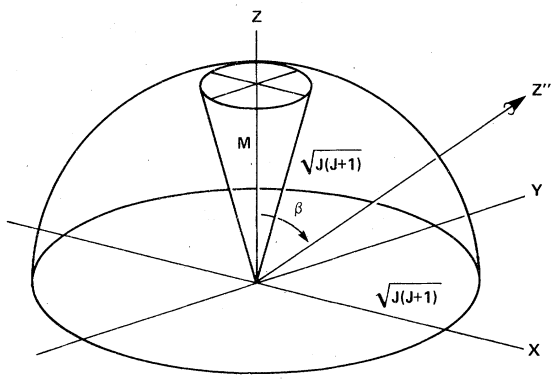

FIG. 3. Representation of the initial population as a cone in abstract space: the cone height is $M=n-\frac{1}{2}(N+1)$ the cone side length is $[J(J+1)]^{1/2}=\frac{1}{2}(N^2-1)^{1/2}$, directed along the Z axis. The vector Z'' shows the axis of the Hamiltonian vector W. Oriented as shown, the cone represents population inversion.

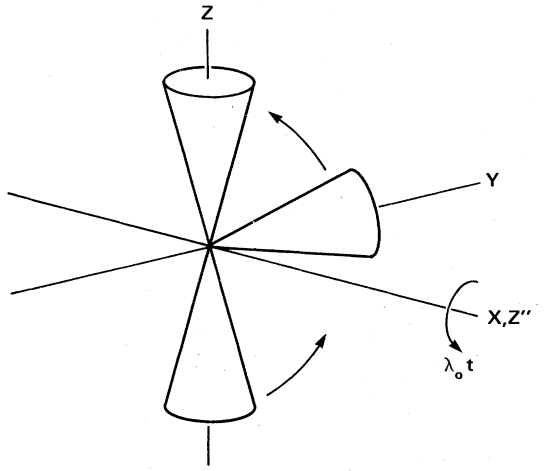

FIG. 4. Representation of population change as a precession of the initial population cone about the axis Z'' of the Hamiltonian vector W. Here there is no detuning or loss and Ω_0 is real, so Z'' coincides with the X axis.

plete inversion. Figure 4 illustrates this progression.

It is interesting to note that if population begins in level n of an N-level atom, then the π pulse places all of the population into level $N-n$ (see Fig. 5). No other levels ever contain all of the population at one instant in time. It is easy to understand this very striking periodic flopping between levels n and $N-n$ from the vector model: 90° rotation of a cone whose height is $+M$, corresponding to level n, produces a cone of height $-M$, corresponding to level $N-n$. Note that when N is an odd integer population confined at $t=0$ to the middle level never accumulates entirely in any other level.

IV. DETUNING

The spin-J model permits cumulative detunings which increase linearly with level number: $\Delta_n = \Delta_0 n$. Such detunings correspond to an idealization in which each laser is detuned by the same frequency, as would occur with Doppler shifts for lasers of very nearly equal frequencies. The detunings shorten the period of population oscillation, in the same way as occurs for the two-level atom: The frequency λ_0 is the rms value of Ω_0 and Δ_0.

The presence of detuning shifts the body axis of the vector W toward the Z axis. This means a corresponding tilt toward the vertical of the precession axis of the population cone for the vector model. When this occurs complete inversion no longer takes place. As we readily visualize from the vector model, an increase in detuning forces the population oscillations to remain within a decreasing band of levels. In the limit of very large detuning (small angle β) the population cone precesses about the Z axis and no population changes occur.

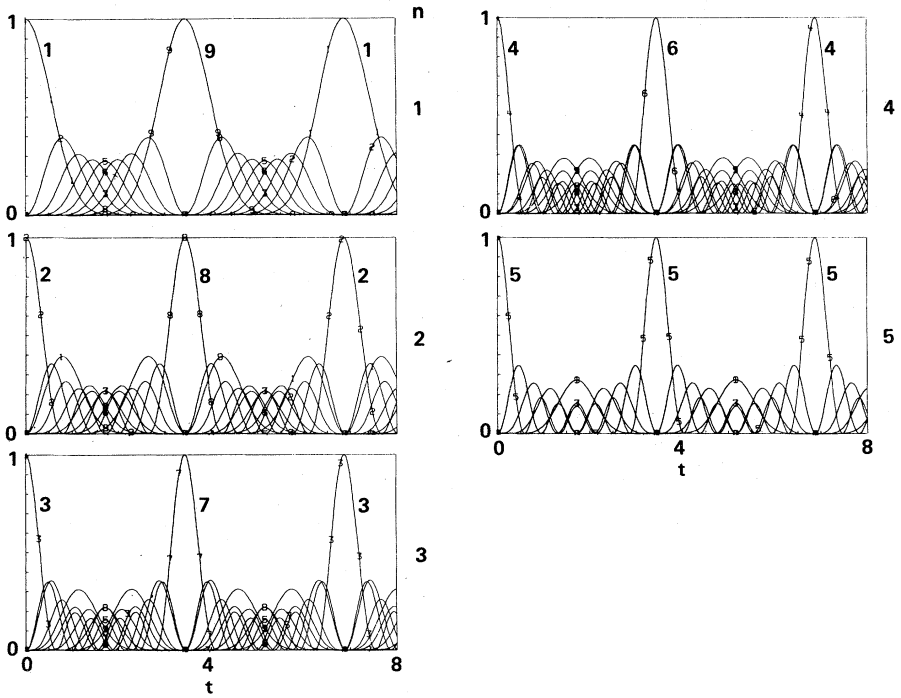

FIG. 5. Effect upon populations $P_n(t)$ for an $N=9$ level atom of varying initial conditions. Successive frames show evolution of system prepared, respectively, in levels 1, 2, 3, 4, and 5. Labels identify curves for specific levels n.

V. IONIZATION

The angular momentum model also allows probability loss proportional to level number: $\gamma_n = \gamma_0 n$. The losses give complex values of the population pulsation period λ_0 leading, in turn, to solutions which decay exponentially with time. The total probability, summed over all N levels, falls exponentially, modulated sinusoidally. The angle β, and thence an argument of the rotation matrix, becomes complex (pure imaginary in the absence of detuning.)

ACKNOWLEDGMENT

This work was performed under the auspices of the U. S. Dept. of Energy at Lawrence Livermore Laboratory under Contract No. W-7405-Eng-48.

[1] A. Einstein, Phys. Z. 18, 121 (1917).
[2] *Selected Papers on Physical Processes in Ionized Plasmas*, edited by D. H. Menzel (Dover, New York, 1962).
[3] V. A. Ambartsumyan, *Theoretical Astrophysics* (Pergamon, New York, 1958).
[4] J. W. Bond, Jr., K. M. Watson, and J. A. Welch, Jr., *Atomic Theory of Gas Dynamics* (Addison-Wesley, Reading, Mass., 1965).
[5] B. W. Shore and M. A. Johnson, J. Chem. Phys. 68, 5631 (1978).
[6] *High-Resolution Laser Spectroscopy*, edited by K. Shimoda (Springer-Verlag, Berlin, 1976).
[7] J. H. Eberly and P. Lambroboulos, *Multiphoton Processes* (Wiley, New York, 1978).
[8] M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, Mass., 1974).
[9] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley-Interscience, New York, 1975).
[10] J. D. Macomber, *The Dynamics of Spectroscopic Transitions* (Wiley, New York, 1976).
[11] G. Oliver, Lett. Nuovo Cimento 2, 1075 (1971).
[12] I. R. Senitzky, Phys. Rev. A 10, 1868 (1974).
[13] D. I. Gruev, Sov. J. Quantum Electron. 5, 1355 (1975).
[14] G. W. F. Drake and R. B. Grimley, Phys. Rev. A 11, 1614 (1975).
[15] R. Gilmore, C. M. Bowden, and L. M. Narducci, Phys. Rev. A 12, 1019 (1975).
[16] A. M. F. Lau, Phys. Rev. A 14, 279 (1976).
[17] T. H. Einwohner, J. Wong, and J. C. Garrison, Phys. Rev. A 14, 1452 (1976).
[18] J. Wong, J. C. Garrison, and T. H. Einwohner, Phys. Rev. A 16, 213 (1977).
[19] G. Oliver, Phys. Rev. A 15, 2323 (1977).
[20] D. M. Larsen, Opt. Commun. 19, 404 (1976).
[21] J. T. Hougen, J. Chem. Phys. 65, 1035 (1976).
[22] D. P. Hodgkinson and J. S. Briggs, Chem. Phys. Lett. 43, 451 (1976).
[23] D. M. Larsen and N. Bloembergen, Opt. Commun. 17, 254 (1976).
[24] J. Stone, M. F. Goodman, and D. A. Dows, J. Chem. Phys. 65, 5062 (1976).
[25] M. F. Goodman, J. Stone, and D. A. Dows, J. Chem. Phys. 65, 5052 (1976).
[26] S. Mukamel and J. Jortner, J. Chem. Phys. 65, 5204 (1976).
[27] D. P. Hodgkinson and J. S. Briggs, J. Phys. B 10, 7583 (1977).
[28] L. M. Narducci, S. S. Mitra, R. A. Shatas, and C. A. Coulter, Phys. Rev. A 16, 247 (1977).
[29] V. S. Letokhov and A. A. Makarov, Opt. Commun. 17, 250 (1976).
[30] M. Sargent III and P. Horwitz, Phys. Rev. A 13, 1962 (1976).
[31] B. W. Shore and J. Ackerhalt, Phys. Rev. A 15, 1640 (1977).
[32] J. H. Eberly, B. W. Shore, Z. Bialynicka-Birula, and I. Bialynicki-Birula, Phys. Rev. A 16, 2038 (1977).
[33] Z. Bialynicka-Birula, I. Bialynicki-Birula, J. H. Eberly, and B. W. Shore, Phys. Rev. A 16, 2048 (1977).
[34] A. A. Makarov, Zh. Eksp. Teor. Fiz. 72, 1749 (1977) [Sov. Phys. JETP 45, 918 (1977)].
[35] B. W. Shore and J. H. Eberly, Opt. Commun. 24, 83 (1978).
[36] A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University, Princeton, N.J., 1957).
[37] R. P. Feynman, F. L. Vernon, Jr., and R. W. Hellwarth, J. Appl. Phys. 28, 49 (1957).