

Coherent population trapping in N -level quantum systems

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A multilevel quantum system interacting with an intense laser field is shown to exhibit many invariants, or constants of evolution, under various conditions. Our results also apply to other similar physical problems.

I. INTRODUCTION

The problem of coherent population trapping and more generally of dynamic symmetry when a multilevel quantum system is irradiated by lasers has received increased attention in recent years.¹⁻¹³ Simply put, dynamic symmetry implies, in addition to the conservation of the total atomic population, one or more other constants of evolution in the dynamics of the system. The problem is to find such invariants, and the conditions under which the system would possess such invariants. Of particular interest is the case when the N -level system has the SU(2) dynamic symmetry,¹² in which the dynamics of the system simplifies because it allows the breakdown of SU(N) symmetry into its O(3) subgroup, and the case when the N -level system has the Gell-Mann dynamic symmetry,^{7,13} in which the breakdown of SU(N) symmetry into its SU(2) and $N-2$ U(1) subgroups is the characteristic feature. The conditions needed for these symmetries to occur and the constants of evolution which arise are given in the references cited.

In this paper, we shall present a number of other conditions under which a multilevel quantum system irradiated by lasers may possess important invariants. In particular, we shall examine a number of cases under the two-photon, three-photon, and generally r -photon resonance conditions. We shall present the constants of evolution in each case.

II. N -LEVEL SYSTEM AT TWO-PHOTON RESONANCE

We consider an N -level or N -state quantum system at two-photon resonance whose generally time-dependent Hamiltonian can be written in or reduced into the form given by

$$\hat{H}(t) = -\hbar \begin{pmatrix} 0 & \alpha_{12}(t) & 0 & \alpha_{14}(t) & \cdots \\ \alpha_{21}(t) & \Delta_2(t) & \alpha_{23}(t) & 0 & \\ 0 & \alpha_{32}(t) & 0 & \alpha_{34}(t) & \\ \alpha_{41}(t) & 0 & \alpha_{43}(t) & \Delta_4(t) & \\ 0 & \alpha_{52}(t) & 0 & \alpha_{54}(t) & \\ \vdots & & & & \ddots \end{pmatrix}. \quad (2.1)$$

The ground state of the system is level 1, and the other levels are so labeled that

$$\alpha_{jk}(t) = 0 \text{ if } |j-k| \text{ is an even number,} \quad (2.2)$$

according to the electric dipole transition rule. In the language of laser physics, the nonzero off-diagonal element $\alpha_{jk}(t)$ is the half-Rabi frequency associated with the transition from level j to k . We shall refer to the off-diagonal elements generally as the interaction parameters. The diagonal element $\Delta_k(t)$ is the cumulative detuning of $k-1$ successive lasers from the corresponding sum of $k-1$ level frequencies. The zero in the first diagonal element only expresses the fact that we can add to or subtract from $\hat{H}(t)$ any multiple of the unit matrix. The subsequent zeros along the diagonal at odd places, or $\Delta_k(t) = 0$ when k is odd, are the result of the two-photon resonance assumption. We assume that $\hat{H}(t)$ is Hermitian and hence

$$\alpha_{kj}(t) = \alpha_{jk}^*(t). \quad (2.3)$$

We further assume, as we do for all the cases we discuss in this paper, that the Rabi frequencies for the allowed transitions have the form

$$\alpha_{jk}(t) = \begin{cases} \alpha_{jk} f(t) & \text{for } |j-k| \text{ an odd number and } j \text{ odd} \\ \alpha_{jk} f^*(t) & \text{for } |j-k| \text{ an odd number and } j \text{ even,} \end{cases} \quad (2.4a)$$

$$(2.4b)$$

where α_{jk} are arbitrary generally complex constants and $f(t)$ is an arbitrary time-dependent function. Note that $f(t)$ and $f^*(t)$ appear in odd and even rows, respectively, of the Hamiltonian matrix. In the simpler case when $f(t)$ is a real function of time, Eq. (2.4) implies that all the Rabi frequencies have the same time dependence. If all the allowed transitions between the levels are derived from the same laser, then Eq. (2.4) is automatically satisfied. Also, if all the Rabi frequencies are constants independent of the time, then Eq. (2.4) is satisfied automatically.

The time evolution of the system, in the time period short compared to the natural decay times, is specified by the N components $\Psi_j(t)$, $j = 1, 2, \dots, N$ of the wave vec-

tor $\Psi(t)$ which satisfies the time-dependent Schrödinger equation

$$i\hbar \frac{\partial \Psi}{\partial t} = \hat{H}(t)\Psi. \quad (2.5)$$

The population of level j at time t is given by $|\Psi_j(t)|^2$, and the conservation of the total population is expressed by

$$|\Psi(t)|^2 = \sum_{j=1}^N |\Psi_j(t)|^2 = 1. \quad (2.6)$$

We shall see that for an N -level system at two-photon resonance whose Hamiltonian is expressed by Eq. (2.1), no matter what the interaction parameters α_{jk} and the one-photon detuning functions $\Delta_{2n}(t)$ are, there is already another invariant if N is odd.

To see this, let us first denote the matrix obtained from $\hat{H}(t)$ by setting all the diagonal elements equal to zero and setting the common time-dependent factors $f(t)$ and $f^*(t)$ equal to 1, by \hat{H}_0 , i.e.,

$$\hat{H}_0 = \begin{pmatrix} 0 & \alpha_{12} & 0 & \alpha_{14} & \cdots \\ \alpha_{21} & 0 & \alpha_{23} & 0 & \\ 0 & \alpha_{32} & 0 & \alpha_{34} & \\ \alpha_{41} & 0 & \alpha_{43} & 0 & \\ 0 & \alpha_{52} & 0 & \alpha_{54} & \\ \vdots & & & & \ddots \end{pmatrix}. \quad (2.7)$$

Odd N implies that the determinant of \hat{H}_0 is zero, hence it also implies that an eigenvalue of \hat{H}_0 is zero. Let the corresponding eigenvector be of the form

$$\mathbf{u}_0 = \begin{pmatrix} k_1 \\ 0 \\ k_3 \\ 0 \\ k_5 \\ \vdots \\ k_N \end{pmatrix}, \quad (2.8)$$

where k_1, k_3, \dots, k_N can be calculated from the even rows of \hat{H}_0 , i.e., from

$$\begin{aligned} \alpha_{21}k_1 + \alpha_{23}k_3 + \alpha_{25}k_5 + \cdots + \alpha_{2N}k_N &= 0, \\ \alpha_{41}k_1 + \alpha_{43}k_3 + \alpha_{45}k_5 + \cdots + \alpha_{4N}k_N &= 0, \\ &\vdots \end{aligned} \quad (2.9)$$

$$\alpha_{N-1,1}k_1 + \alpha_{N-1,3}k_3 + \alpha_{N-1,5}k_5 + \cdots + \alpha_{N-1,N}k_N = 0.$$

Note that we have $\frac{1}{2}(N-1)$ conditions to determine $\frac{1}{2}(N+1)$ constants. At least one of the k 's is assumed to be nonzero from this set of equations, and we shall express the remaining k 's in terms of it. For example, if k_1 is not zero, then let us write Eq. (2.9) as

$$\hat{A}\mathbf{k} = -k_1\mathbf{a}_1, \quad (2.10)$$

where

$$\hat{A} = \begin{pmatrix} \alpha_{23} & \alpha_{25} & \alpha_{27} & \cdots & \alpha_{2N} \\ \alpha_{43} & \alpha_{45} & \alpha_{47} & \cdots & \alpha_{4N} \\ \vdots & & & & \\ \alpha_{N-1,3} & \alpha_{N-1,5} & \alpha_{N-1,7} & \cdots & \alpha_{N-1,N} \end{pmatrix}, \quad (2.11)$$

$$\mathbf{k} = \begin{pmatrix} k_3 \\ k_5 \\ k_7 \\ \vdots \\ k_N \end{pmatrix}, \quad \mathbf{a}_1 = \begin{pmatrix} \alpha_{21} \\ \alpha_{41} \\ \alpha_{61} \\ \vdots \\ \alpha_{N-1,1} \end{pmatrix}. \quad (2.12)$$

Let $D = \det \hat{A}$, $D_{2j+1} = \det \hat{A}_{2j+1}$, $j = 1, 2, \dots, (N-1)/2$, where \hat{A}_{2j+1} is the matrix obtained from \hat{A} by replacing the column $(\alpha_{2,2j+1}, \alpha_{4,2j+1}, \dots, \alpha_{N-1,2j+1})$ by \mathbf{a}_1 . Then we have

$$k_{2j+1} = -(D_{2j+1}/D)k_1, \quad j = 1, 2, \dots, (N-1)/2 \quad (2.13)$$

which determine all the components of the eigenvector (2.8) corresponding to the zero eigenvalue of \hat{H}_0 . It is easy to see that $\hat{H}(t)\mathbf{u}_0$ gives a zero column vector, and if \mathbf{u}_0^\dagger denotes a row vector which is the complex-conjugate transpose of \mathbf{u}_0 , then $\mathbf{u}_0^\dagger \hat{H}(t)$ gives a zero row vector, i.e., $\hat{H}_0\mathbf{u}_0 = 0$ and $\mathbf{u}_0^\dagger \hat{H}_0 = 0$. Thus, multiplying Eq. (2.5) from the left by \mathbf{u}_0^\dagger , we find

$$\frac{\partial}{\partial t} [\mathbf{u}_0^\dagger \Psi(t)] = 0 \quad (2.14)$$

or

$$\Psi_1(t) - \sum_{j=1}^{(N-1)/2} \frac{D_{2j+1}^*}{D^*} \Psi_{2j+1}(t) = \text{const}. \quad (2.15)$$

We have thus shown that for a multilevel system with an odd number of levels N which interacts with a laser field under the two-photon resonance condition, there is a second constant of evolution given by Eq. (2.15) besides the obvious one given by Eq. (2.6) in the dynamics of the system. Equation (2.15) expresses a coherent population trapping which is present when the system of an odd number of levels is operated at the two-photon resonance condition. For $N=3$, Eq. (2.15) gives

$$\Psi_1(t) - \frac{\alpha_{12}}{\alpha_{23}^*} \Psi_3(t) = \text{const}, \quad (2.16)$$

which coincides with the constant of evolution discovered by Gray, Whitley, and Stroud,² and coincides also with a characteristic constant of motion when the system possesses the so-called Gell-Mann dynamic symmetry.^{7,13} For $N > 3$, Eq. (2.15) is new.

If the number of levels N of the system is even, then \hat{H}_0 does not in general have a zero eigenvalue and Eq. (2.15) does not apply. There is no obvious second constant of evolution unless other conditions are imposed, as we shall discuss this in Sec. III.

III. TWO-PHOTON RESONANCE AND EQUAL ONE-PHOTON DETUNINGS

We next consider the case when the one-photon detunings $\Delta_{2n}(t)$ in (2.1) are equal at all times, i.e.,

$$\Delta_2(t) = \Delta_4(t) = \cdots \equiv \Delta(t). \quad (3.1)$$

Thus we assume the condition of two-photon resonance and equal one-photon detunings. If $\Delta(t) = 0$, then we have the special case of one-photon resonance at all times.

We will show that under condition (3.1), there is a unitary transformation by a time-independent unitary matrix \hat{U} such that the transformed Hamiltonian $\hat{\mathcal{H}}(t)$ obtained from

$$\hat{\mathcal{H}}(t) = \hat{U}^\dagger \hat{H}(t) \hat{U} \quad (3.2)$$

is a block diagonal matrix of the form

$$\hat{\mathcal{H}}(t) = -\hbar \begin{pmatrix} 0 & h_{12}(t) & 0 & 0 & 0 & 0 \\ h_{21}(t) & \Delta(t) & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & h_{34}(t) & 0 & 0 \\ 0 & 0 & h_{43}(t) & \Delta(t) & 0 & 0 \\ 0 & 0 & 0 & 0 & \ddots & \\ 0 & 0 & 0 & 0 & & \ddots \end{pmatrix}, \quad (3.3)$$

where one of the diagonal blocks is a single zero if N is odd.

We shall prove the above assertion in several steps. First we notice that if λ is an eigenvalue of \hat{H}_0 of Eq. (2.7), then $-\lambda$ is also an eigenvalue of \hat{H}_0 . To see this, we write the equation to determine the components k_j of an eigenvector of \hat{H}_0 as

$$\sum_n \alpha_{jn} k_n = \lambda k_j, \quad j = 1, 2, \dots, N \quad (3.4)$$

where the summation on the left is over all even values of n if j is odd, and is over all odd values of n if j is even. If we can solve these equations for an eigenvalue and eigenvector, we get another solution by changing the signs of λ and the odd components.

The following two vectors \mathbf{u}_p^- and \mathbf{u}_p^+ formed by computing the difference and the sum of the two eigenvectors corresponding to the eigenvalues $\pm\lambda_p$ are convenient basis states which we shall use:

$$\mathbf{u}_p^- \equiv \begin{pmatrix} k_1^{(p)} \\ 0 \\ k_3^{(p)} \\ 0 \\ k_5^{(p)} \\ 0 \\ \vdots \end{pmatrix}, \quad \mathbf{u}_p^+ \equiv \begin{pmatrix} 0 \\ k_2^{(p)} \\ 0 \\ k_4^{(p)} \\ 0 \\ k_6^{(p)} \\ \vdots \end{pmatrix}. \quad (3.5)$$

It is easy to verify that

$$\begin{aligned} \hat{H}_0 \mathbf{u}_p^- &= \lambda_p \mathbf{u}_p^+, \\ \hat{H}_0 \mathbf{u}_p^+ &= \lambda_p \mathbf{u}_p^-. \end{aligned} \quad (3.6)$$

We now form a unitary matrix \hat{U} whose columns consist of orthonormalized \mathbf{u}_p^- and \mathbf{u}_p^+ , $p = 1, 2, \dots, [N/2]$, where $[N/2]$ denotes the integer part of $N/2$. If N is odd, the last column is the eigenvector \mathbf{u}_0 corresponding to the zero eigenvalue of \hat{H}_0 . It can now be verified that when (3.1) holds, the unitary transformation (3.2) gives the result (3.3), where

$$h_{j,j+1}(t) = \lambda_p f(t) \quad (3.7a)$$

and

$$h_{j+1,j}(t) = \lambda_p f^*(t) = h_{j,j+1}^*(t). \quad (3.7b)$$

We have thus reduced the dynamics of the N -level system operated under the condition of two-photon resonance and equal one-photon detunings into that of $[N/2]$ two-level systems whose time evolutions are given by

$$i\hbar \frac{\partial}{\partial t} \begin{pmatrix} \Psi'_{1p} \\ \Psi'_{2p} \end{pmatrix} = -\hbar \begin{pmatrix} 0 & \lambda_p f(t) \\ \lambda_p f^*(t) & \Delta(t) \end{pmatrix} \begin{pmatrix} \Psi'_{1p} \\ \Psi'_{2p} \end{pmatrix}, \quad (3.8)$$

where $\pm\lambda_p$, $p = 1, 2, \dots, [N/2]$ are the eigenvalues of \hat{H}_0 given by (2.7), and where

$$\Psi' = \hat{U}^\dagger \Psi \quad (3.9a)$$

or

$$\Psi'_{1p} = \mathbf{u}_p^{-\dagger} \cdot \Psi, \quad \Psi'_{2p} = \mathbf{u}_p^{+\dagger} \cdot \Psi. \quad (3.9b)$$

It follows that we have the following set of constants of evolution:

$$|\mathbf{u}_p^{-\dagger} \cdot \Psi(t)|^2 + |\mathbf{u}_p^{+\dagger} \cdot \Psi(t)|^2 = \text{const}, \quad p = 1, 2, \dots, [N/2] \quad (3.10a)$$

and the following one additional constant if N is odd:

$$\mathbf{u}_0^\dagger \cdot \Psi(t) = \text{const}. \quad (3.10b)$$

In terms of the components of \mathbf{u}_p^\pm given by (3.5), which are the components $k_j^{(p)}$ given by (3.4) of the eigenvectors of \hat{H}_0 corresponding to the eigenvalues $\pm\lambda_p$, and in terms of the components $\Psi_j(t)$ of $\Psi(t)$, Eq. (3.10) can be expressed more explicitly as

$$\left| \sum_{n=1}^{[N/2]} k_{2n-1}^{(p)*} \Psi_{2n-1}(t) \right|^2 + \left| \sum_{n=1}^{[N/2]} k_{2n}^{(p)*} \Psi_{2n}(t) \right|^2 = \text{const}, \quad p = 1, 2, \dots, [N/2] \quad (3.11a)$$

and

$$\sum_{n=1}^{[N/2]} k_{2n-1}^{(0)*} \Psi_{2n-1} = \text{const} \quad \text{for odd } N. \quad (3.11b)$$

The reduction to the equivalent two-level systems (3.8) allows the solution of the N -level system to be expressed analytically for a large number of cases.¹⁴

If all the λ_p equal zero except one, the system is said to possess the Gell-Mann-type dynamic symmetry.⁷ The

condition for and the consequence of this symmetry have been discussed in greater detail in earlier papers.^{7,13}

IV. APPLICATIONS TO $N = 4$ AND 5

For $N = 3$, the result of Sec. III simply reproduces the result previously given. However, beginning with $N = 4$ and up, applications of the results of the preceding sections already give new results, and it is useful to see some specific examples.

Noting our results as expressed by Eqs. (3.11) and (3.8), it is sufficient for us to give the expressions for $k_{2n+1}^{(p)}/k_1^{(p)}$, $k_{2n}^{(p)}/k_2^{(p)}$, and λ_p . For $N = 4$, we find

$$\frac{k_3^{(p)}}{k_1^{(p)}} = \frac{\alpha_{32}\alpha_{21} + \alpha_{34}\alpha_{41}}{\lambda_p^2 - (|\alpha_{23}|^2 + |\alpha_{34}|^2)}, \quad (4.1a)$$

$$\frac{k_4^{(p)}}{k_2^{(p)}} = \frac{\alpha_{41}\alpha_{12} + \alpha_{43}\alpha_{32}}{\lambda_p^2 - (|\alpha_{34}|^2 + |\alpha_{14}|^2)}, \quad (4.1b)$$

$$\lambda_p^2 = \frac{1}{2}[s \pm (s^2 - 4d)^{1/2}], \quad p = 1, 2 \text{ for } +, - \quad (4.1c)$$

where

$$s = |\alpha_{12}|^2 + |\alpha_{23}|^2 + |\alpha_{34}|^2 + |\alpha_{14}|^2, \quad (4.1d)$$

$$d = |\alpha_{12}\alpha_{34} - \alpha_{14}\alpha_{32}|^2. \quad (4.1e)$$

For $N = 5$, we find

$$\frac{k_3^{(p)}}{k_1^{(p)}} = \frac{(\alpha_{21}\alpha_{32} + \alpha_{34}\alpha_{41})\lambda_p^2 - (\alpha_{21}\alpha_{45} - \alpha_{41}\alpha_{25})(\alpha_{32}\alpha_{54} - \alpha_{34}\alpha_{52})}{\lambda_p^4 - (|\alpha_{23}|^2 + |\alpha_{34}|^2 + |\alpha_{45}|^2 + |\alpha_{52}|^2)\lambda_p^2 + |\alpha_{23}\alpha_{45} - \alpha_{43}\alpha_{25}|^2}, \quad (4.2a)$$

$$\frac{k_5^{(p)}}{k_1^{(p)}} = \frac{(\alpha_{21}\alpha_{52} + \alpha_{41}\alpha_{54})\lambda_p^2 - (\alpha_{23}\alpha_{41} - \alpha_{43}\alpha_{21})(\alpha_{32}\alpha_{54} - \alpha_{34}\alpha_{52})}{\lambda_p^4 - (|\alpha_{23}|^2 + |\alpha_{34}|^2 + |\alpha_{45}|^2 + |\alpha_{52}|^2)\lambda_p^2 + |\alpha_{23}\alpha_{45} - \alpha_{43}\alpha_{25}|^2}, \quad (4.2b)$$

$$\frac{k_4^{(p)}}{k_2^{(p)}} = \frac{\alpha_{12}\alpha_{41} + \alpha_{32}\alpha_{43} + \alpha_{45}\alpha_{52}}{\lambda_p^2 - (|\alpha_{34}|^2 + |\alpha_{45}|^2 + |\alpha_{14}|^2)} \text{ for } p = 0, 1, 2, \quad (4.2c)$$

$$\lambda_p^2 = \frac{1}{2}[s \pm (s^2 - 4d)^{1/2}], \quad p = 1, 2 \text{ for } +, - \quad (4.2d)$$

and $\lambda_0 = 0$, where

$$s = |\alpha_{12}|^2 + |\alpha_{23}|^2 + |\alpha_{34}|^2 + |\alpha_{45}|^2 + |\alpha_{14}|^2 + |\alpha_{25}|^2 \quad (4.2e)$$

and

$$d = |\alpha_{12}\alpha_{34} - \alpha_{14}\alpha_{32}|^2 + |\alpha_{12}\alpha_{54} - \alpha_{14}\alpha_{52}|^2 + |\alpha_{23}\alpha_{45} - \alpha_{25}\alpha_{43}|^2. \quad (4.2f)$$

To see how these cases reduce to the cases of Gell-Mann symmetry,^{7,13} let us first consider the case of $N = 4$. We observe from Eq. (4.1c) that if $d = 0$, then two of the eigenvalues of \hat{H}_0 , $\pm\lambda_2$ become zero. Thus the condition for this to occur is, from (4.1e),

$$\alpha_{12}\alpha_{34} = \alpha_{14}\alpha_{32}. \quad (4.3)$$

The two nonzero eigenvalues $\pm\lambda_1$ are given by

$$\lambda_1^2 = |\alpha_{12}|^2 + |\alpha_{23}|^2 + |\alpha_{34}|^2 + |\alpha_{14}|^2. \quad (4.4)$$

The eigenvectors, (4.1a) and (4.1b), simplify to

$$k_3^{(1)}/k_1^{(1)} = \alpha_{32}/\alpha_{12}, \quad k_4^{(1)}/k_2^{(1)} = \alpha_{43}/\alpha_{23}, \quad (4.5)$$

$$k_3^{(2)}/k_1^{(2)} = -\alpha_{21}/\alpha_{23}, \quad k_4^{(2)}/k_2^{(2)} = -\alpha_{32}/\alpha_{34}.$$

The transformed Hamiltonian (3.3) becomes

$$\hat{H}(t) = -\hbar \begin{pmatrix} 0 & \lambda_1 f(t) & 0 & 0 \\ \lambda_1 f^*(t) & \Delta(t) & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \Delta(t) \end{pmatrix}. \quad (4.6)$$

For the case $N = 5$, we observe from Eq. (4.2d) that if $d = 0$, then $\lambda_2^2 = 0$. The condition for this to occur is, from (4.2f)

$$\alpha_{jk} = a_j a_k^*, \quad (4.7)$$

for $|j - k|$ equal to an odd number, where a_j are arbitrary generally complex constants. Equation (2.2) is assumed to hold throughout this paper. The two nonzero eigenvalues are $\pm\lambda_1$ given by

$$\lambda_1^2 = |\alpha_{12}|^2 + |\alpha_{23}|^2 + |\alpha_{34}|^2 + |\alpha_{45}|^2 + |\alpha_{14}|^2 + |\alpha_{25}|^2. \quad (4.8)$$

For the eigenvectors corresponding to the triply degenerate zero eigenvalue, Eq. (4.2c) for $\lambda = 0$ gives an eigenvector whose components are given by

$$k_4 : k_4 = a_4^* : -a_2^*, \quad (4.9)$$

and Eqs. (4.2a) and (4.2b) for $\lambda = 0$ suggest two independent eigenvectors whose components are given, respectively, by

$$k_1 : k_3 : k_5 = a_3^* : -a_1^* : 0 \quad (4.10a)$$

and

$$k_1 : k_3 : k_5 = a_5^* : 0 : -a_1^*. \quad (4.10b)$$

The two eigenvectors given by Eq. (4.10) are not orthogonal but can be made orthogonal by the Gram-Schmidt process. After the eigenvectors are orthonormalized, the transformed Hamiltonian (3.3) becomes

$$\hat{H}(t) = -\hbar \begin{pmatrix} 0 & \lambda_1 f(t) & 0 & 0 & 0 \\ \lambda_1 f^*(t) & \Delta(t) & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \Delta(t) & 0 \\ 0 & 0 & 0 & 0 & 0 \end{pmatrix}, \quad (4.11)$$

where λ_1 is given by Eq. (4.8).

Equation (4.7) allows the generalization to an N -level system for the system to possess the Gell-Mann symmetry.^{7,13} In that case, we have

$$\lambda_1^2 = \sum_{m < n} |\alpha_{mn}|^2 \quad (4.12a)$$

and

$$\lambda_p = 0 \text{ for } p = 2, 3, \dots, [N/2]. \quad (4.12b)$$

Thus from Eqs. (3.8)–(3.10), we have

$$|\Psi'_{1p}|^2 + |\Psi'_{2p}|^2 = \text{const for } p = 1, \quad (4.13a)$$

$$\Psi'_{1p}(t) = \Psi'_{1p}(0), \quad (4.13b)$$

$$\Psi'_{2p}(t) = \Psi'_{2p}(0) \exp \left[i \int \Delta dt \right] \quad \text{for } p = 2, 3, \dots, [N/2], \quad (4.13c)$$

or, more explicitly, the characteristic constants of evolution are

$$\left| \sum_{n=1}^{[N/2]} k_{2n-1}^{(p)*} \Psi_{2n-1}(t) \right|^2 + \left| \sum_{n=1}^{[N/2]} k_{2n}^{(p)*} \Psi_{2n}(t) \right|^2 = \text{const for } p = 1, \quad (4.14a)$$

$$\sum_{n=1}^{[N/2]} k_{2n-1}^{(p)*} \Psi_{2n-1}(t) = \text{const} \quad \text{for } p = 2, 3, \dots, [N/2] \text{ and } p = 0 \text{ if } N \text{ is odd,} \quad (4.14b)$$

$$\left| \sum_{n=1}^{[N/2]} k_{2n}^{(p)*} \Psi_{2n}(t) \right| = \text{const for } p = 2, 3, \dots, [N/2], \quad (4.14c)$$

where $k_j^{(p)}$ are components of the orthonormalized eigenvectors corresponding to the eigenvalues $\pm \lambda_p$.

V. CHAINWISE DIPOLE CONNECTED N -LEVEL SYSTEMS

We consider in this section a special class of N -level systems whose levels are principally only chainwise dipole connected, i.e.,

$$\alpha_{jk} = \alpha_{kj}^* = 0 \text{ unless } |j - k| = 1, \quad (5.1)$$

and for N an even number, we assume also that

$$\alpha_{1N} = \alpha_{N1}^* \neq 0, \quad (5.2)$$

generally, which is allowed by the dipole selection rule. Thus for even N , the dipole connections complete a loop, whereas for odd N , the dipole connections complete only a chain. All the other dipole connections are assumed to be negligibly small. These cases are not only of importance in practice, but the results given in Sec. II and III can also be expressed more simply and explicitly. We shall express some of these results below.

(i) At two-photon resonance and N odd. Condition (5.1) sets all the off-diagonal elements in the Hamiltonian (2.1) equal to zero except those immediately above and below the diagonal. The constant of evolution given by Eq. (2.15) can be written more explicitly in this case as

$$\Psi_1(t) - \frac{\alpha_{12}}{\alpha_{23}^*} \Psi_3(t) + \frac{\alpha_{12}\alpha_{34}}{\alpha_{23}^*\alpha_{45}^*} \Psi_5(t) - \frac{\alpha_{12}\alpha_{34}\alpha_{56}}{\alpha_{23}^*\alpha_{45}^*\alpha_{67}^*} \Psi_7(t) + \dots + (-1)^{(N-1)/2} \frac{\alpha_{12}\alpha_{34}\alpha_{56} \dots \alpha_{N-2,N-1}}{\alpha_{23}^*\alpha_{45}^*\alpha_{67}^* \dots \alpha_{N-1,N}^*} \Psi_N(t) = \text{const}. \quad (5.3)$$

(ii) Two-photon resonance with equal one-photon detunings and N even. The nonzero off-diagonal elements of the Hamiltonian $\hat{H}(t)$ consist of those immediately above and below the diagonal, and two more, α_{1N} and α_{N1} , at the corners. The nonzero diagonal elements in (2.1) are assumed to be all equal to $\Delta(t)$. The determinant of \hat{H}_0 of (2.7) is in this case given by

$$\det \hat{H}_0 = (-1)^{N/2} |\alpha_{12}\alpha_{34}\alpha_{56} \dots \alpha_{N-1,N} + (-1)^{(N/2)-1} \alpha_{32}\alpha_{54}\alpha_{76} \dots \alpha_{1N}|^2. \quad (5.4)$$

If $\det \hat{H}_0 = 0$, i.e., if

$$\alpha_{12}\alpha_{34}\alpha_{56} \dots \alpha_{N-1,N} = (-1)^{N/2} \alpha_{32}\alpha_{54}\alpha_{76} \dots \alpha_{1N}, \quad (5.5)$$

then \hat{H}_0 has at least a pair of zero eigenvalues which results in a pair of constants of evolution linear in $\Psi_j(t)$ given by

$$\Psi_1(t) - \frac{\alpha_{12}}{\alpha_{23}^*} \Psi_3(t) + \frac{\alpha_{12}\alpha_{34}}{\alpha_{23}^*\alpha_{45}^*} \Psi_5(t) - \frac{\alpha_{12}\alpha_{34}\alpha_{56}}{\alpha_{23}^*\alpha_{45}^*\alpha_{67}^*} \Psi_7(t) + \dots + (-1)^{(N/2)-1} \frac{\alpha_{12}\alpha_{34}\alpha_{56} \dots \alpha_{N-3,N-2}}{\alpha_{23}^*\alpha_{45}^*\alpha_{67}^* \dots \alpha_{N-2,N-1}^*} \Psi_{N-1}(t) = \text{const} \quad (5.6a)$$

and

$$\left| \Psi_2(t) - \frac{\alpha_{23}}{\alpha_{34}^*} \Psi_4(t) + \frac{\alpha_{23}\alpha_{45}}{\alpha_{34}^*\alpha_{56}^*} \Psi_6(t) - \frac{\alpha_{23}\alpha_{45}\alpha_{67}}{\alpha_{34}^*\alpha_{56}^*\alpha_{78}^*} \Psi_8(t) \right. \\ \left. + \cdots + (-1)^{(N/2)-1} \frac{\alpha_{23}\alpha_{45}\alpha_{67} \cdots \alpha_{N-2,N-1}}{\alpha_{34}^*\alpha_{56}^*\alpha_{78}^* \cdots \alpha_{N-1,N}^*} \Psi_N(t) \right| = \text{const} . \quad (5.6b)$$

VI. N -LEVEL SYSTEM AT r -PHOTON RESONANCE, AND r IS EVEN AND GREATER THAN 2

Instead of the two-photon resonance condition as we have assumed in all the previous cases, suppose that the N -level system is at r -photon resonance between levels 1 and $r+1$, r is even, and $2 < r < N$. The off-diagonal elements of the Hamiltonian $\hat{H}(t)$ are assumed to be as in Eq. (2.1), but the elements along the diagonal are now of the form $[0, \Delta_2(t), \Delta_3(t), \dots, \Delta_r(t), 0, \Delta_{r+2}(t), \dots, \Delta_N(t)]$, where, except for the first element and the element at the $(r+1)$ th position which are zeros, the other elements are generally not zero and may assume any arbitrary functions of time.

We wish to construct a vector \mathbf{u}_0 such that

$$\hat{H}(t)\mathbf{u}_0 = 0 . \quad (6.1)$$

Let \mathbf{u}_0 be a column vector which has only two nonzero components at the first and the $(r+1)$ th positions, i.e.,

$$\mathbf{u}_0 = \begin{pmatrix} k_1 \\ 0 \\ 0 \\ \vdots \\ 0 \\ k_{r+1} \\ 0 \\ \vdots \\ 0 \end{pmatrix} , \quad (6.2)$$

where k_1 and k_{r+1} are constants. Then Eq. (6.1) could be satisfied if

$$\frac{\alpha_{21}}{\alpha_{2,r+1}} = \frac{\alpha_{41}}{\alpha_{4,r+1}} = \frac{\alpha_{61}}{\alpha_{6,r+1}} = \cdots = \frac{\alpha_{N-1,1}}{\alpha_{N-1,r+1}} = -\frac{k_{r+1}}{k_1} \quad (6.3)$$

Thus, provided that the off-diagonal elements of $\hat{H}(t)$ satisfy Eq. (6.3), multiplying both sides of Eq. (2.5) by \mathbf{u}_0^\dagger , a row vector which is the complex-conjugate transpose of \mathbf{u}_0 of (6.2) whose two nonzero components satisfy (6.3), gives the following second constant of evolution for the system at r -photon resonance:

$$\alpha_{2,r+1}^* \Psi_1(t) - \alpha_{21} \Psi_{r+1}^*(t) = \text{const} . \quad (6.4)$$

A similar analysis can be applied to an r -photon resonance condition between any two levels of an N -level system.

VII. SUMMARY

We have found and presented a number of constants of evolution in the dynamics of a multilevel quantum system interacting with an intense laser field, and the conditions under which such invariants can be expected. The principal results are given by (i) Eq. (2.15) when the N -level system is operated at two-photon resonance, and N is odd, (ii) Eqs. (3.10) or (3.11) and (3.8) when the system is operated at two-photon resonance and equal one-photon detunings, (iii) the results in Secs. IV and V for some special cases, and (iv) Eq. (6.4) when the system is operated at an r -photon resonance between levels 1 and $r+1$. The invariants are characteristics of the dynamics of the system under the specified conditions which can be usefully exploited in practice.

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¹E. Arimondo and G. Orriols, *Nuovo Cimento Lett.* **17**, 333 (1976).

²H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., *Opt. Lett.* **3**, 218 (1978).

³F. J. Cook and B. W. Shore, *Phys. Rev. A* **20**, 539 (1979).

⁴J. D. Stettler, C. M. Bowden, N. M. Witroil, and J. H. Eberly, *Phys. Lett.* **73A**, 171 (1979).

⁵F. T. Hioe and J. H. Eberly, *Phys. Rev. A* **25**, 2168 (1982).

⁶P. M. Radmore and P. L. Knight, *J. Phys. B* **15**, 561 (1982).

⁷F. T. Hioe, *Phys. Rev. A* **28**, 879 (1983); **32**, 2824 (1985).

⁸Z. Deng, *Opt. Commun.* **48**, 284 (1983).

⁹F. T. Hioe, *Phys. Rev. A* **29**, 3434 (1984); **30**, 3097 (1984).

¹⁰H. P. W. Gottlieb, *Phys. Rev. A* **26**, 3713 (1982); **32**, 653 (1985); D. T. Pegg, *J. Phys. B* **18**, 415 (1985).

¹¹S. J. Buckle, S. M. Barnett, P. L. Knight, M. A. Lauder, and D. T. Pegg, *Opt. Acta* **33**, 1129 (1986).

¹²F. T. Hioe, *J. Opt. Soc. Am. B* **4**, 1327 (1987). The (3,1) element for $j=1$ in Eq. (16) of this reference should be $+b^{*2}$, not $-b^{*2}$.

¹³F. T. Hioe, *J. Opt. Soc. Am. B* (to be published).

¹⁴F. T. Hioe and C. E. Carroll, *Phys. Rev. A* **32**, 1541 (1985); C. E. Carroll and F. T. Hioe, *J. Phys. Rev. A* **19**, 3579 (1986).