Reduction of degenerate two-level excitation to independent two-state systems

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Examining the dynamics of a degenerate two-level system excited by coherent resonant light, we exhibit a transformation of basis states which reduces the problem to a set of independent nondegenerate one- and two-state systems. More generally, reduction of multilevel Hamiltonians requires only that the states can be collected into two sets; within each set the states are degenerate, unlinked by Hamiltonian matrix elements. The remaining Hamiltonian linkages must share the same time dependence, but are otherwise arbitrary. We apply this method to reduction of the angular momentum degeneracy of a two-level atom excited by elliptically polarized light and to multiple-laser resonant excitation of multiple levels into a common level.

INTRODUCTION

The nondegenerate two-level atom (or, more precisely, two-state atom) provides an important model in treating time-dependent excitation (see Allen and Eberly¹) and serves as the basic starting point for more elaborate studies of atomic and molecular excitation by lasers. The presence of magnetic sublevels can complicate the appearance of the dynamical equations,²⁻⁷ but in this paper we show that, even with arbitrary elliptically polarized light and with degenerate sublevels, the coherently excited nearresonant decay-free two-level atom can be treated as a set of independent two-state systems. (Of course, this superposition of two-state systems does not behave as any effective "average" two-state system.)

Figure 1 illustrates the reduction of interest. Figure 1(a) shows the dipole-transition linkages present with elliptically polarized excitation (a combination of $\Delta M = +1$ and -1, where M is the magnetic quantum number) for transitions between levels having angular momenta J=2 and 1. In the absence of radiative decay, the dynamics comprises two independent systems: a three-state " λ " (dashed line) and a five-state M (solid line). We show that one can always introduce a new basis of upper- and lower-level states (replacing the basis of magnetic sublevels) within which the dynamics follows a pattern appropriate to the one- and two-state linkages of Fig. 1(c). The linkage patterns of Figs. 1(a) and 1(c) are familiar from expressions of linear polarization in terms of a quantization axis along the propagation direction [Fig. 1(a)] or along the electric vector [Fig. 1(c)]. The present, more general, transformation applies to excitation with arbitrary multipolarity and polarization as well as to arbitrary degeneracy.

Although we term the system of interest a degenerate two-level atom, we point out below that transformation applies to a class of more general multilevel systems.



FIG. 1. (a) Excitation linkage pattern for electricdipole transitions between a ground level having angular momentum J=2 and an excited level having angular momentum J=1. The pattern comprises two separate linkages, an M system shown as full lines, and a λ shown as dashed lines. Labels show magnetic quantum number M. (b) System dynamically equivalent to that of (a), obtained by introducing new degenerate basis states for the M linkage. (c) System dynamically equivalent to that of (a), obtained by introducing new basis states for the Mand the λ systems.

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STATEMENT OF THE PROBLEM

The problem of interest requires solution of a time-dependent Schrödinger equation

$$i\frac{d}{dt}c_i(t) = \sum_i H_{ij}c_j(t)$$

for the probability amplitudes $c_i(t)$. The Hamiltonian matrix H has the structure

$$[H] = \begin{bmatrix} h_1 & V \\ V^{\dagger} & h_2 \end{bmatrix},$$

where h_1 and h_2 are diagonal submatrices and where V denotes the linkages between sublevels of level 1 (having angular momentum J_1) and those of level 2 (angular momentum J_2) originating in the laser driven excitation.

In the rotating-wave approximation (RWA) the elements h_i represent detunings, and V is a matrix of (slowly varying) Rabi frequencies, proportional to the electric field strength and to appropriate Clebsch-Gordan coefficients. For our method we only need assume that h_1 and h_2 are (possibly time-dependent) scalar multiples of unit matrices and that the ratio of any two elements of V is time independent.

Now we can always introduce a unitary transformation on the basis states

$$c' = Uc$$

with

$$U^{\dagger}U = 1 = UU^{\dagger}$$

such that in the new basis the amplitudes satisfy the matrix equation

$$i\frac{d}{dt}c' = H'c$$

with

$$H' = UHU^{\dagger}$$
.

This is equivalent to introducing a new set of basis states $|i\rangle'$

$$|i\rangle' = \sum_{j} |j\rangle U_{ji}^{\dagger}$$
.

We shall show that we can always find a transformation such that H' represents independent oneand two-state transitions.

THE TRANSFORMATION

The transformation of interest must only mix sublevels of a given level. Thus we can express the matrix U in terms of unitary matrices A and B

$$\begin{bmatrix} U \end{bmatrix} = \begin{bmatrix} A & 0 \\ 0 & B \end{bmatrix}$$
 with

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$$A^{\dagger}A = I_1 = AA^{\dagger}, B^{\dagger}B = I_2 = BB^{\dagger}$$

Here I_1 and I_2 are the identity matrices appropriate to the dimensions $2J_1 + 1$ and $2J_2 + 1$, respectively. Then the transformed Hamiltonian has the form

$$[H'] = \begin{bmatrix} h'_1 & M \\ M^{\dagger} & h'_2 \end{bmatrix}$$

where

$$h'_1 = Ah_1A^{\dagger}$$
, $h'_2 = Bh_2B^{\dagger}$, $M = AVB^{\dagger}$

Although the matrices h_i and A and B are square matrices, the matrices M and V need not be; levels 1 and 2 may have different degeneracies. Our goal is to obtain a matrix M for which each sublevel of the lower state connects to no more than one upper sublevel and vice versa. Thus if we extract from rectangular M a square matrix \overline{M} , then \overline{M} will be a diagonal matrix when the sublevels are suitably ordered. The eigenvalues of \overline{M} are half the Rabi frequencies for the transformed problem.

If we are to succeed in representing the degenerate two-level atom as independent two-state systems, then the (square) matrices h'_i must be diagonal. The original matrices h_i are, by the degeneracy supposition, scalar multiples of the identity matrices I_i . (Indeed, they are null matrices for exactly resonant excitation.) Thus the transformed matrices h'_i will indeed be diagonal, and furthermore, they will maintain the original degeneracy; we have

 $h_i' = h_i$.

The assumption of degeneracy rules out application to strong magnetic fields or appreciable hyperfine splitting whenever the sublevel energy spacing becomes a non-negligible fraction of the nonzero elements of V.

Next we observe that

$$MM^{\dagger} \equiv AVV^{\dagger}A^{\dagger}$$
 and $M^{\dagger}M \equiv BV^{\dagger}VB^{\dagger}$

are square matrices. As a step toward our goal of a diagonal matrix \overline{M} , let us require that these be diagonal. That is, we define (in part) the unitary matrices A and B by requiring that they diagonalize VV^{\dagger} and $V^{\dagger}V$, respectively. Now the eigenvalues of VV^{\dagger} and $V^{\dagger}V$ are real and non-negative, so that we can write them as squares. To prove this assertion we note that if u is a non-null eigenvector of VV^{\dagger} ,

$$VV^{\mathsf{T}}u = \lambda u$$
,

then

$$u^{\dagger}(VV^{\dagger}u) = \lambda ||u||^2$$

where ||u|| denotes the norm of u. But we also have

$$u^{\dagger}(VV^{\dagger}u) = (V^{\dagger}u)^{\dagger}(V^{\dagger}u) = ||V^{\dagger}u||^{2} \ge 0$$

Thus the eigenvalue λ must be real and non-negative.

We therefore can write the diagonalized matrices in the form

$$AVV^{\dagger}A^{\dagger} = MM^{\dagger} = a^2$$
,
 $BV^{\dagger}VB^{\dagger} = M^{\dagger}M = b^2$

(that is, the matrix a^2 has diagonal elements a_i^2). We require that the new basis states are ordered such that elements of a^2 and b^2 are arranged in nonincreasing order. (The number a_i is half the Rabi frequency for a two-state transition.) If each of the matrices a^2 and b^2 consists of distinct nonzero diagonal elements, then we can identify the desired matrix \overline{M} as $(a^2)^{1/2}$ or $(b^2)^{1/2}$, since both a^2 and b^2 will then be square and equal to each other (as shown below). However, the general case will admit degeneracies and null eigenvalues with which we must deal.

Next, we want to eliminate the null subspaces of the matrices V and V^{\dagger} . [By definition these yield null values for elements of $M = AVB^{\dagger}$, and hence they correspond to (uninteresting) unlinked levels which do not participate in the excitation dynamics.] We proceed by removing all of the null eigenvalues of a^2 and b^2 to form the (diagonal) matrices a^2 and b^2 ,

$$a^2 \rightarrow \overline{a}^2$$
 and $b^2 \rightarrow \overline{b}^2$.

Similarly, we discard the corresponding rows of A, Band columns of A^{\dagger}, B^{\dagger} to form new matrices $\overline{A}, \overline{B}$,

$$A \to \overline{A}$$
 and $B \to \overline{B}$.

Using these truncated matrices we then construct the desired matrix M,

$$\overline{M} = \overline{A} V \overline{B}^{\dagger}$$
.

Now according to our construction we have

$$\overline{M}\,\overline{M}^{\dagger} = \overline{a}^{\,2}$$
 and $\overline{M}^{\dagger}\,\overline{M} = \overline{b}^{\,2}$

We can show that, except for their null spaces, MM^{\dagger} and $M^{\dagger}M$ have equal eigenvalues of equal multiplicities. Let v_j be an eigenvector of MM^{\dagger} having a nonzero eigenvalue

$$MM^{\dagger}v_j = a_j^2 v_j$$

with

$$a_j^2 \neq 0$$
,

and let u_i be the vector

$$u_i = M^{\dagger} v_i$$
.

Then

$$M^{\dagger}Mu_{j} = M^{\dagger}(MM^{\dagger})v_{j} = a_{j}^{2}M^{\dagger}v_{j} = a_{j}^{2}u_{j}$$
,

and u_j is an eigenvector of $M^{\dagger}M$ having eigenvalue a_j^2 . The vectors $\{u_j\}$ are linearly independent (otherwise some combination of $\{v_j\}$ would be in the excluded null space of MM^{\dagger}). Thus we see that $M^{\dagger}M$ has the same eigenvalues and multiplicities as MM^{\dagger} . Because \bar{a}^2 and \bar{b}^2 are diagonal with eigenvalues arranged in nondecreasing order, they are equal. Hence \overline{M} commutes with its adjoint

$$\overline{M}\,\overline{M}^{\dagger} = \overline{M}^{\dagger}\,\overline{M}$$
,

so that \overline{M} is a normal matrix.⁸

Now any normal matrix can be diagonalized by a unitary transformation.⁸ We can therefore prove that, because the matrix \overline{M} is normal and $\overline{M} \overline{M}^{\dagger}$ is diagonal, \overline{M} can be decomposed into the product of a diagonal matrix and a block unitary matrix Q (see the Appendix). The diagonal factor is just the matrix $\overline{a} = \overline{b}$, where all the diagonal elements are chosen to be real and positive. Thus we can write

$$M=\bar{a}Q$$
,

where Q is block unitary,

$$Q^{\dagger}Q = QQ^{\dagger} = I_3$$

and where the matrix of interaction strengths is the diagonal matrix

$$\bar{a} = (\bar{A} V V^{\dagger} \bar{A}^{\dagger})^{1/2}$$

Here I_3 is the unit matrix appropriate to the dimensionality of \overline{M} . If the matrix \overline{a}^2 is nondegenerate, then Q will be the unit matrix; if eigenvalues of \overline{a} occur in pairs, then Q will comprise 2×2 blocks.

Given the matrix \overline{M} (which we obtain by constructing $\overline{A}V\overline{B}^{\dagger}$), we can obtain the matrix Q by dividing by the diagonal matrix \overline{a} ,

$$Q = (\overline{a})^{-1}\overline{M}$$
 or $Q^{\dagger} = \overline{M}^{\dagger}(\overline{a})^{-1}$

Because \bar{a} has, by construction, no null eigenvalues, the inverse $(\bar{a})^{-1}$ is well defined. We can now use this matrix to construct a transformation for the level-2 sublevels

$$\widetilde{B}^{\dagger} = \overline{B}^{\dagger}Q^{\dagger} = \overline{B}^{\dagger}\overline{B}V^{\dagger}A^{\dagger}(\overline{a})^{-1}$$

which will complete the diagonalization of the interaction V. That is, the matrix

$$\widetilde{M} = \overline{A} V \widetilde{B}^{\dagger}$$

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is diagonal. It has the elements

$$\widetilde{M} = \overline{a} = (\overline{A} V V^{\dagger} \overline{A}^{\dagger})^{1/2}$$

(Note that although $\overline{B} \overline{B}^{\dagger} = I_2$, the reverse is not true, $\overline{B} \,\overline{B}^{\dagger} \neq \overline{B}^{\dagger} \,\overline{B}$.) The desired transformation of the Hamiltonian therefore has the form

$$[U] = \begin{vmatrix} \overline{A} & 0 \\ A^0 & 0 \\ 0 & \overline{B} \\ 0 & B^0 \end{vmatrix},$$

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where A^0 and B^0 denote the null spaces of A and B.

EXAMPLE

To clarify how this diagonalization proceeds in practice we examine a particular case. Consider the example of the "M" linkage portion of the RWA Hamiltonian of the J=2 to 1 excitation shown in Fig. 1(a). The elements of the RWA Hamiltonian within levels are

$$h_1 = \omega_1 I_1 = \omega_1 \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix},$$
$$h_2 = \omega_2 I_2 = \omega_2 \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}.$$

We are free to choose the RWA phase such that $\omega_1 = 0$, and h_1 is a null matrix. Then ω_2 expresses the detuning of the laser from the Bohr transition frequency; h_2 will be null if the laser is tuned to resonance.

To evaluate the interaction operator V we employ an helicity basis with states labeled by magnetic quantum number M appropriate to quantization along the propagation direction. We express the electric-dipole interaction operator in the form

$$\hat{V} = \left[\left[\frac{1+p}{2} \right]^{1/2} \hat{d}_{-1}x + \left[\frac{1-p}{2} \right]^{1/2} \hat{d}_{+1}x^* \right] \epsilon , \qquad \text{This transformation is eq} \\ \text{basis states from those labe number } |M\rangle \text{ to states } |n\rangle \\ |1\rangle' = [|-1\rangle x (f+5p)^{1/2} + |+1\rangle x^* (f-5p)^{1/2}] / \sqrt{2f} \xrightarrow[p=0]{} (|+1\rangle + |-1\rangle) / \sqrt{2} \\ \stackrel{p=0}{\xrightarrow[p=\pm 1]} |\mp 1\rangle , \\ |2\rangle' = [-|-1\rangle x (f-5p)^{1/2} + |+1\rangle x^* (f+5p)^{1/2}] / \sqrt{2f} \xrightarrow[p=0]{} (-|-1\rangle + |+1\rangle) \\ \stackrel{p=\pm 1}{\xrightarrow[p=\pm 1]} \stackrel{p=0}{\xrightarrow[p=\pm 1]} \pm |\pm 1\rangle .$$

Thus pure circular polarization $(p = \pm 1)$ corresponds, at most, to a phase change. For linear polarization (p=0) these states $|1\rangle'$ and $|2\rangle'$ together with the third state

$$|3\rangle' = |0\rangle$$

(part of the λ subsystem of states for the $J=2\rightarrow J=1$ system) form, apart from phase factors, a real irreduci-

where ϵ is the electric field strength, \hat{d}_q is the dipole moment operator, $x = \exp(i\varphi)$ is a phase factor, and the parameter p, bounded by $-1 \le p \le +1$, expresses polarization: $p = \pm 1$ for circular, p = 0 for linear. Within the M system the RWA matrix V has the form

$$V = \frac{\Omega}{\sqrt{20}} \begin{bmatrix} x[6(1+p)]^{1/2} & 0\\ x^*(1-p)^{1/2} & x(1+p)^{1/2}\\ 0 & x^*[6(1-p)]^{1/2} \end{bmatrix} \begin{pmatrix} (-2)\\ (0)\\ (+2) \end{pmatrix}$$

where Ω is the real-valued Rabi frequency.

To find the transformed basis states we begin with the excited level because it has the smaller degeneracy, two. We form the matrix

$$V^{\dagger}V = \frac{\Omega^2}{20} \begin{bmatrix} 7+5p & x^{2}(1-p^2)^{1/2} \\ (x^*)^{2}(1-p^2)^{1/2} & 7-5p \end{bmatrix}$$

The transformation **B** which diagonalizes $V^{\dagger}V$ is

$$B = \frac{1}{\sqrt{2f}} \begin{bmatrix} x^{*}(f+5p)^{1/2} & x(f-5p)^{1/2} \\ -x^{*}(f-5p)^{1/2} & x(f+5p)^{1/2} \end{bmatrix},$$

where

$$f \equiv (1 + 24p^2)^{1/2}$$

The eigenmatrix b^2 is $BV^{\dagger}VB^{\dagger}$, so that the matrix b is

$$b = \frac{\Omega}{\sqrt{20}} \begin{bmatrix} (7+f)^{1/2} & 0\\ 0 & (7-f)^{1/2} \end{bmatrix}.$$

The two eigenvalues are distinct and nonzero (because $-1 \le p \le 1$ and thus $1 \le f \le 5$) so we have

$$\overline{B} = B$$
 and $\overline{b} = b$.

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ble tensor set⁹—as opposed to a standard magnetic sublevel basis in a rotated coordinate system.

To find the basis states for the ground level we next form the matrix

$$VV^{\dagger} = \frac{\Omega^2}{20} \begin{vmatrix} 6(1+p) & x^2 [6(1-p^2)]^{1/2} & 0\\ (x^*)^2 [6(1-p^2)]^{1/2} & 2 & x^2 [6(1-p^2)]^{1/2}\\ 0 & (x^*)^2 [6(1-p^2)]^{1/2} & 6(1-p) \end{vmatrix} .$$

By diagonalizing this matrix we obtain three eigenvalues, one of which is zero. The nonzero eigenvalues generate the transformation

$$\bar{A} = \begin{bmatrix} \frac{(x^*)^2}{2}(1+p)(6p-1+f)N_+ & \sqrt{6}p(1-p^2)^{1/2}N_+ & \frac{x^2}{2}(1-p)(6p+1-f)N_+ \\ \frac{(x^*)^2}{2}(1+p)(6p-1-f)N_- & \sqrt{6}p(1-p^2)^{1/2}N_- & \frac{x^2}{2}(1-p)(6p+1+f)N_- \end{bmatrix}$$

where

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$$N_{\pm} = \{f[(1+p^2)f \mp (1-11p^2)]\}^{-1/2}$$

ſ

and the complementary null space generates the transformation

$$A^{0} = \{-(x^{*})^{2}(1-p) \quad [6(1-p^{2})]^{1/2} \quad -x^{2}(1+p)]\}/2(2-p^{2})^{1/2}$$

The matrix \overline{M} is then obtainable as the product

$$\overline{M} = \overline{A}V\overline{B}^{\dagger} = \frac{\Omega}{\sqrt{20}} \begin{bmatrix} (7+f)^{1/2} & 0\\ 0 & (7-f)^{1/2} \end{bmatrix}.$$

In this case \overline{M} is completely diagonal; it is identical with the matrix $\overline{a} = \overline{b}$. Thus the block-unitary matrix Q is just the unit matrix, and in this example

$$\widetilde{B} = \overline{B}$$
 and $\widetilde{M} = \overline{M}$.

We see that, in this example, the upper-level transformation $\tilde{B} = \bar{B} = B$ mixes the two sublevels having magnetic quantum number |M| = 1 to produce two new states. The lower-level transformation \bar{A} mixes the sublevels having |M| = 0,2 to produce two states, each linked with a unique one of these upper states, and one unlinked state which does not participate in the excitation. That is, we have the Hamiltonian excitation pattern of Fig. 1(b) (the λ linkage remains unaffected by this transformation).

Our transformation does not alter the detuning; resonant excitation remains resonant. The elements of \overline{M} are half the Rabi frequencies for the two transitions of the new two-level systems; these are never identical.

To complete the treatment of the $J=2\rightarrow J=1$ system we need to carry out a similar (and wellknown) transformation of the λ portion of Figs. 1(a) and 1(b); this results in the pattern of Fig. 1(c), in which we obtain three two-level excitation linkages and two unlinked lower states. In the case of linear polarization we can identify the transformed basis states as those appropriate to a real irreducible tensor set, rather than the standard magnetic sublevel basis.

COMMENTS

We have shown that we can reduce the degenerate two-level system to independent two-state systems through the following prescription: First, we obtain unitary matrices A and B which diagonalize VV^{\dagger} and $V^{\dagger}V$,

$$AVV^{\dagger}A^{\dagger} = a^2$$
 and $BV^{\dagger}VB^{\dagger} = b^2$

We then remove the null subspaces to obtain matrices \overline{A} and \overline{B} . Finally, we form the matrix \widetilde{B}^{\dagger}

$$\widetilde{B}^{\dagger} = \overline{B}^{\dagger} \overline{B} V^{\dagger} \overline{A}^{\dagger} (\overline{a})^{-1}$$

Then the new basis states are

$$|i\rangle' = \sum_{j} \overline{A}_{ji}^{\dagger} |j\rangle$$

for level 1, and
 $|i\rangle' = \sum_{j} \widetilde{B}_{ji}^{\dagger} |j\rangle$

for level 2. The nonzero linkages (i.e., Rabi frequencies) between these new basis states are the elements \bar{a} . The transformation of basis states does not alter the resonant nature of the excitation; the matrices h_i remain null matrices after the transformation.

The resulting dynamical simplification can be considerable. Instead of dealing with an $N \times N$ Hamiltonian matrix, where $N = (2J_1+1)$ $+(2J_2+1)$, we need only deal with *n* independent two-state systems, where *n* is at most the lesser of $2J_1+1$ and $2J_2+1$. In general, each two-state system has a different Rabi flopping frequency, so that the overall population dynamics is not a simple sinusoidal Rabi flopping.

We note that both of the transformations A and B

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(from which we obtain \overline{A} and \overline{B}) depend upon the linkage pattern between levels 1 and 2 rather than depending upon properties of the levels alone, for both diagonalize a bilinear form VV^{\dagger} or $V^{\dagger}V$. In any attempt to construct a multilevel ladder using a sequence of transformations A, B, C, D, \ldots , all but the first and last transformations must simultaneously diagonalize two quadratic forms. Only specially related pairs of quadratic forms can be diagonalized simultaneously by a single unitary transformation. Thus the general degenerate multilevel excitation cannot always be reduced to independent ladders.

The two-state reduction discussed in the present paper differs from the reduction of a sequentially linked multilevel system to an equivalent two-level system which occurs when intermediate levels are not resonant or when intermediate Rabi frequencies are very large.¹⁰

To construct our transformation it is not necessary that the interaction matrix V be independent of time. It is only necessary that the time dependence be a common factor in all elements; otherwise the basis-state expansion changes with time. Of course, it is always possible, in principle, to choose sufficiently small time intervals that ratios of matrix elements remain constant; different basis states then apply within each interval.

Although the foregoing discussion dealt with linkage patterns arising from excitation of degenerate magnetic sublevels by polarized light, the proof of reduction to a two-state system made no use of the rotational symmetry properties of the interaction V. In particular, it does not require that the elements of V be related in any way to angular momentum operators, although such is of course the case for the example treated above. Furthermore, the matrices h_i need not be real (and hence Hermitian), but may include an imaginary term to express probability loss due to ionization or dissociation. Thus our analysis applies to the well-known λ system^{11,12} of Fig. 2(a) and to the other multibranched linkages of Fig. 2(b). In these systems a set of n distinct lasers excites a set of n lower levels into a common upper level. Each laser is resonantly tuned to the appropriate transition frequency, so that in the RWA¹³ the elements of h_i are degenerate. (Recall that in the RWA the elements of h_i are the detunings; the RWA dynamics is independent of the energies.) We have shown that such systems are dynamically equivalent to the system of Fig. 2(c), in which only a single lower sublevel connects with the excited state. Thus if the ground sublevels initially have equal populations, then only 1/n of the ground-level population can ever reach the excited state and subsequently ionize. The remainder of the population



FIG. 2. (a) λ system linkage pattern for two resonantly tuned lasers. (b) Multileg linkage pattern. (c) Equivalent system to (b).

remains trapped in a coherent superposition of ground levels. If the lasers are detuned from resonance, the elements of h_i no longer are equal (to zero), and the lack of degeneracy prevents transformation to a two-state system; the ionized fraction can exceed 1/n. Alternatively, if different lasers have different temporal behavior, then we no longer have the conditions necessary for application of our transformation (i.e., time-independent ratios of V-matrix elements), and again the ionized fraction can exceed 1/n. (Sequential access can yield complete ionization.)

In this paper we have shown that certain classes of coherent-excitation linkage patterns reduce to independent two-state excitations. When this occurs the coherent-excitation relaxation-free equations of motion for an appropriate $N \times N$ density matrix reduce by transformation to unlinked equations for 2×2 and 1×1 density matrices.

The considered patterns are those for which the linked states can be collected into two sets (or levels); within each set the states are degenerate in the RWA. That is, no off-diagonal matrix elements link states of a common level. There is no further restriction upon the linkage pattern. The only restriction on values of matrix elements is that the ratio of any two be time independent. Barring accidental zeros, the number of 2×2 systems is the dimensionality of the smaller set of states, i.e., the smaller of the two-level degeneracies.

The present paper aims only to show the existence of this transformation and to point out simple applications to excitation of degenerate two-level systems by elliptically polarized light (of arbitrary multipolarites) and to excitation of multibranched generalizations of the well-known λ system. It would be interesting to examine the algebraic or grouptheoretical properties of the transformation coefficients themselves for cases in which angular momentum operators yield the elements of the interaction matrix and for which tensorial sets⁹ offer a popular basis for expressing the density matrix.

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APPENDIX

We have the following. Theorem: If M is a normal matrix and if

$$(MM^{\dagger})_{ij} = a_i^2 \delta_{ij} \tag{A1}$$

with

 $a_i \ge a_i > 0$

for i > j, then $a^{-1}M$ is block unitary in blocks that are identical to the degenerate blocks of a.

Proof: From Eq. (A1) we obtain the result

 $a^{-1}MM^{\dagger}a^{-1}=I_3$,

- ¹L. Allen and J. H. Eberly, *Optical Resonance and Two-*Level Atoms (Wiley, New York, 1975).
- ²B. A. Zon and B. G. Katsnel'son, Zh. Eksp. Teor. Fiz. 65, 947 (1973) [Sov. Phys.—JETP 38, 470 (1974)].
- ³A. L. Burshtein, É. G. Saprykin, and G. L. Smirnov, Zh. Eksp. Teor. Fiz. <u>66</u>, 1570 (1974) [Sov. Phys.—JETP <u>39</u>, 769 (1974)].
- ⁴B. W. Shore, Phys. Rev. A <u>17</u>, 1739 (1978).
- ⁵M. Aihara, Phys. Rev. A <u>18</u>, 606 (1978).
- ⁶A. Crubellier and M. G. Schweighofer, Phys. Rev. A <u>18</u>, 1797 (1978).
- ⁷J. Cooper, R. J. Ballagh, and K. Burnett, Phys. Rev. A 22, 535 (1980).
- ⁸P. R. Halmos, Finite Dimensional Vector Spaces (Van Nostrand, Princeton, New Jersey, 1958), p. 159; J. H. Wilkinson, The Algebraic Eigenvalue Problem (Oxford University Press, Oxford, 1965), p. 51.
- ⁹A. Omont, Prog. Quantum Electron. <u>5</u>, 80 (1977).
- ¹⁰B. W. Shore, Phys. Rev. A <u>24</u>, 1413 (1981).

so that

$$(a^{-1}M)(a^{-1}M)^{\dagger}=I_{1}$$

and the matrix $a^{-1}M$ is unitary. Again from Eq. (A1) and from $M^{\dagger}M = MM^{\dagger}$ we have the results

$$M^{\dagger} = a^2 M^{-1} = M^{-1} a^2$$
,

so that a^2 commutes with M,

 $[a^2, M] = 0$.

From this result we have the result

$$0 = a[a,M] + [a,M]a$$
,

so that for all i and j

$$0 = [a, M]_{ij}(a_i + a_j)$$

or, because $a_i > 0$ for all i,

 $0 = [a, M]_{ii}$.

Therefore *a* commutes with $a^{-1}M$,

$$0 = [a, a^{-1}M]$$

and, for all i and j,

 $0 = (a_i - a_i)(a^{-1}M)_{ii}$

so that, as was to be proved, the matrix $a^{-1}M$ has elements only within blocks sharing a common eigenvalue.

- ¹¹R. M. Whitley and C. R. Stroud, Jr., Phys. Rev. A <u>14</u>, 1498 (1976); H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. <u>3</u>, 218 (1978); C. R. Stroud, Jr. and D. A. Cardimona, Opt. Commun. <u>37</u>, 221 (1981).
- ¹²W. E. Bell and A. L. Bloom, Phys. Rev. Lett. <u>6</u>, 623 (1961); E. Arimondo and G. Orriols, Lett. Nuovo Cimento <u>17</u>, 333 (1976); K. Kolwas and M. Kolwas, Opt. Commun. <u>17</u>, 149 (1976); B. W. Shore, Laser Program Annual Report No. UCRL-50021-76, Lawrence Livermore National Laboratory, 1976 (unpublished), p. 8; D. P. Hodginson and J. S. Briggs, Opt. Commun. <u>22</u>, 45 (1977); G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento <u>36B</u>, 5 (1977); G. Orriols, *ibid*. <u>53B</u>, 1 (1979); D. F. Walls and P. Zoller, Opt. Commun. <u>34</u>, 260 (1980).
- ¹³T. H. Einwohner, J. Wong, and J. C. Garrison, Phys. Rev. A <u>14</u>, 1452 (1976); Z. Bialynicka-Birula, I. Bialynicki-Birula, J. H. Eberly, and B. W. Shore, *ibid*. <u>16</u>, 2048 (1977).

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