# Coherent excitation of a degenerate two-level system by an elliptically polarized laser pulse

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The elaborate linkage pattern of the interaction Hamiltonian descriptive of pulsed laser interaction of elliptically polarized light with a degenerate two-level system can, for coherent excitation, be reduced to sets of independent pairs of coupled two-state equations by means of the Morris-Shore transformation [Phys. Rev. A **27**, 906 (1983)]. By extending the earlier defining work, which dealt only with time-independent interactions, to consider various pulse shapes, one can obtain exact analytic solutions to various multilevel linkages. We use these to find the conditions on the interaction parameters (e.g., the ellipticity) leading to various population transfer schemes, for example, to achieve either population inversion or orientation. We illustrate these with the *M* and *W* linkages amongst magnetic sublevels of $J=1$ or $J=2$ excited by elliptically polarized light.

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## I. INTRODUCTION

The properties of idealized nondegenerate two-state atoms acted on by pulsed laser radiation form a substantial part of the now-standard textbook descriptions of time dependence in quantum mechanics [1,2]. The behavior of *degenerate* two-level systems, such as occurs with systems possessing angular momentum (as characterized by quantum numbers *J* and *M* for degeneracy $2J+1$), requires somewhat more elaborate treatment, including averages over initially populated sublevels (e.g., the magnetic sublevels distinguished by different *M* for given *J*). In the very simplest idealization, that of coherent excitation with linearly polarized light whose direction coincides with the quantization axis used to define the magnetic quantum number *M*, it is only necessary to consider averaging the behavior of sets of independent two-state systems. However, for general choices of quantization axis, or for elliptically polarized light, the laser-interaction Hamiltonian has elements linking more than two quantum states, and the treatment becomes more complicated [3].

The objectives also become more diverse. One may wish to remove all population from all of the lower-lying sublevels, or to put all population into some (superposition of) excited sublevels, or just to redistribute the population amongst the magnetic sublevels (leading to orientation or alignment). For all such tasks it is desirable to have analytic expressions for the time dependence of the various sublevel populations, as will be provided in the present paper.

As discussed in detail by Fewell [3] and others [4–6], the typical electric-dipole interaction Hamiltonian has matrix elements (in an angular-momentum basis associated with quantum numbers *J* and *M*) proportional to Clebsch-Gordon coefficients or $3j$ symbols, and these prescribe not only chains of couplings but also various branched linkage patterns: starting from a particular *M* value, dipole transitions generally induce transitions to states with magnetic quantum numbers $M-1$, *M*, and $M+1$. Hyperfine structure introduces still further linkages [5]. These linking matrix elements of the Hamiltonian produce, for incoherent excitation, transition rates [3] and, for coherent excitation, Rabi frequencies [4]. A consequence of such linkages is that, even in a two-level system (with degenerate sublevels), the task of simulating the response to pulsed radiation can become complicated and no longer amenable to analytic solution.

Fewell [3] has examined examples of degenerate two-level systems and has suggested that some simplification occurs if one is able to choose the polarization appropriately. Here we reconsider this problem—the modeling of excitation by elliptically polarized light—from a different perspective, one that allows the use of the many known exact analytic solutions to two-state coherent excitation by pulsed (and possibly chirped) laser radiation. Our approach is less general than that of Fewell, who allowed arbitrary directions for the quantization axis: we choose this axis along the propagation direction, and express the polarization in terms of the two helicity states associated with this direction.

Some years ago it was recognized by Morris and Shore (MS) [7] that this replacement of a linkage pattern involving two sets of arbitrarily coupled states could be generalized to a variety of multilevel systems, including rather complicated linkages. Under appropriate conditions, reviewed below, it is possible to replace an *N*-state system, described by a constant Hamiltonian matrix, by a set of independent two-state systems. The needed mathematical transformation is a generalization of the change, when dealing with transitions between magnetic sublevels of a system possessing angular momentum, from a helicity basis (left and right circular polarization) to a quantization axis along the electric vector of linear polarization.

The MS factorization has been used in techniques designed to measure the parameters of fully or partly coherent superpositions of two [8] or more [9] states. A similar change of basis states is found in treatments of the $\Lambda$ linkage of three states, such as occuring with stimulated Raman transitions leading to the occurrence of so-called dark states [10,11]. More recently a four-state tripod-linkage system was analyzed using two coupled states and two dark states [12].

 

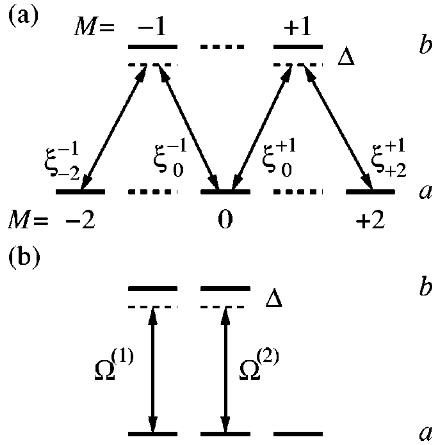


FIG. 1. Frame (a): the linkage pattern of the five-state $M$ system. The states with $M=-2,0,2$ ($\psi_{-2}$, $\psi_0$, and $\psi_2$) form the ground-state manifold, whereas the states with $M=-1,1$ ($\psi_{-1}$ and $\psi_1$) form the excited-state manifold. The coupling pulse is elliptically polarized and the relative coupling strengths, i.e., the Clebsch-Gordon coefficients, are denoted by $\xi_{M_a}^{M_b}$. The pulse is detuned by $\Delta$ from exact resonance. Frame (b): the MS transformation casts this system into a set of two two-state systems and a decoupled state. The driven two-state systems have Rabi frequencies $\Omega^{(i)}$.

The MS factorization has important advantages: rather than dealing with an $N\times N$ Hamiltonian matrix, one has only several separate and independent $2\times 2$ Hamiltonians to solve. Because there are many known analytic solutions to the two-state problem, this approach enables us to find analytic solutions for multistate problems. The present paper therefore extends and generalizes earlier results for a three-state $\Lambda$ system [13]. Such solutions, which allow not only time-varying pulse envelopes but also time-dependent detunings, enable us to derive analytically numerous properties of degenerate two-level systems driven by elliptically polarized laser pulses.

The transformation to decoupled two-state systems is noteworthy of itself, but in addition there are some interesting aspects of the physics. Just as in the $\Lambda$ system, coherence plays a role that is easily overlooked: although the two-state systems can be solved independently, their probability amplitudes must be superposed coherently to obtain the probabilities of physical interest. This superposition introduces constructive and destructive interference, with attendant novel population dynamics.

This paper is organized as follows. The MS transformation is reviewed in Sec. II. In Sec. III we describe an approach based on the MS decomposition, which allows one to find analytic solutions to multistate dynamics by using analytically soluble two-state models. Section IV illustrates this approach for five-state chains with particular application to two cases of magnetic sublevel degeneracy. The population dynamics of a $M$ system (cf. Fig. 1) is explored in Sec. V and that of a $W$ system (cf. Fig. 2) is examined in Sec. VI. Finally, the conclusions are summarized in Sec. VII.

## II. THE MS TRANSFORMATION

We assume completely coherent evolution, i.e., there are no decoherence processes during the interaction. Then the

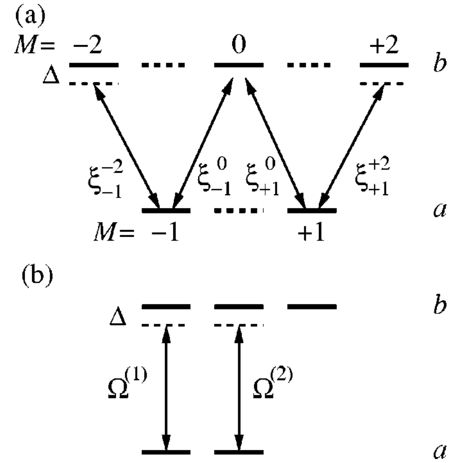


FIG. 2. Same as Fig. 1 but for a $W$-shaped linkage pattern, which can be viewed as an inverted $M$ system. Now the decoupled state is a superposition of excited sublevels.

excitation dynamics is described by the Schrödinger equation, which in the rotating-wave picture and with the rotating-wave approximation (RWA) [4] takes the form of coupled ordinary differential equations for time-dependent complex-valued probability amplitudes $C_n(t)$,

$$\frac{d}{dt}C_n(t)=-i\sum_m W_{nm}C_m(t), \tag{2.1}$$

from which one evaluates the probability of finding the population in state $n$ at time $t$ as $P_n(t)=|C_n(t)|^2$.

The coefficient matrix $\mathsf{W}$, obtained from the Hamiltonian matrix by rescaling from energy to frequency with $\hbar$, and making the RWA, has detunings $\Delta_n=W_{nn}$ as diagonal elements and Rabi frequencies $\Omega_{nm}=2W_{nm}$ as off-diagonal elements (originating typically with electric-dipole interactions).

Though not explicitly shown, the matrix elements may vary with time. For the present interest, only two different detunings $\Delta_n$ occur in the Hamiltonian $\hbar\mathsf{W}$; each may vary (independently) with time. We also assume that all the Rabi frequencies $\Omega_{nm}$ have the same time dependence, although their maximal amplitudes may differ. The special case when all nonzero elements of the Hamiltonian, Rabi frequencies and detunings, have the same time dependence, say $f(t)$, reduces to that of a constant Hamiltonian by defining a new time variable $x=\int f(t)dt$. In the present paper, we do not make this latter assumption and we show that analytic solutions can be found in the more general case when the detunings vary independently of the Rabi frequencies. This includes the important special case of constant detunings and pulse-shaped Rabi frequencies.

With the assumed two distinct detunings, and suitable ordering of the basis states, the matrix $\mathsf{W}$ has the form

$$\mathsf{W}=\begin{bmatrix}\Delta_a\mathsf{I}_a & \mathsf{V}\\ \mathsf{V}^\dagger & \Delta_b\mathsf{I}_b\end{bmatrix}. \tag{2.2}$$

Here $\mathsf{I}_a$ and $\mathsf{I}_b$ are unit matrices, of dimensions $N_a$ and $N_b$, respectively, and $\mathsf{V}$ is a $N_a \times N_b$ matrix of Rabi frequencies (all with the same time dependence). By suitable choice of overall phase for the probability amplitudes one of the two detunings can always be set to zero; this is traditionally taken to be the first of these detunings $\Delta_a$, which is associated with the initially populated state. When all transitions are resonant, as is often assumed, both detunings are zero.

Morris and Shore [7] showed that any such system can be transformed, via suitable redefinition of basis states, to one involving a set of $N_<$ independent two-state systems, where $N_<$ is the lesser of $N_a$ and $N_b$, together with a set of decoupled *spectator* states that are unaffected by the radiation (one-state systems). That is, one is led to equations for new MS amplitudes $\overline{C}_n(t)$,

$$\overline{C}_n(t) = \sum_m U_{nm} C_m(t), \tag{2.3}$$

of the form

$$\frac{d}{dt}\overline{C}_n(t) = -i \sum_m \overline{W}_{nm} \overline{C}_m(t), \tag{2.4}$$

where the matrix $\overline{\mathsf{W}} = \mathsf{U}\mathsf{W}\mathsf{U}^\dagger$ is block diagonal

$$\overline{\mathsf{W}} = \begin{bmatrix} \mathsf{w}^{(1)} & 0 & 0 & \cdots \\ 0 & \mathsf{w}^{(2)} & 0 & \cdots \\ 0 & 0 & \mathsf{w}^{(3)} & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{bmatrix}. \tag{2.5}$$

Though not shown explicitly, the elements of $\mathsf{U}$ and $\overline{\mathsf{W}}$, like those of $\mathsf{W}$ and $\mathsf{w}^{(m)}$, may be time dependent. Here each $\mathsf{w}^{(m)}$ is a one- or two-dimensional matrix. The two-dimensional ones (there are $N_<$ of these) have the form

$$\mathsf{w}^{(m)} = \begin{bmatrix} \Delta_a & \frac{1}{2}\Omega^{(m)} \\ \frac{1}{2}\Omega^{(m)} & \Delta_b \end{bmatrix}, \tag{2.6}$$

where $\Delta_a$ and $\Delta_b$ are the detunings of the original system. The remaining matrices $w^{(m)}$ are one-dimensional (there are $N_> - N_<$ of these). Their elements are detunings, either $\Delta_a$ if $N_a > N_b$, or $\Delta_b$ if $N_b > N_a$. These one-dimensional Hamiltonians induce only time-dependent phase factors $e^{-i\Delta_a t}$ or $e^{-i\Delta_b t}$ in the evolution of the dark-state amplitudes.

The elements of the transformation matrix $\mathsf{U}$ are obtainable from the transformation that diagonalizes the product of interaction matrices $\mathsf{V}\mathsf{V}^\dagger$ by means of a transformation matrix $\mathsf{A}$, which operates in the lower-states manifold (referred to as $A$ space),

$$\mathsf{A}\mathsf{V}\mathsf{V}^\dagger\mathsf{A}^\dagger = \text{diag}, \tag{2.7}$$

and the similar transformation of $\mathsf{V}^\dagger\mathsf{V}$ within the excited states ($B$ space) by means of a matrix $\mathsf{B}$,

$$\mathsf{B}\mathsf{V}^\dagger\mathsf{V}\mathsf{B}^\dagger = \text{diag}. \tag{2.8}$$

In each case the diagonal elements are squares of Rabi frequencies. Given the matrices $\mathsf{A}$ and $\mathsf{B}$, one constructs the desired transformation of the Hamiltonian as

$$\mathsf{U} = \mathsf{G}\begin{bmatrix} \mathsf{A} & 0 \\ 0 & \mathsf{B} \end{bmatrix}\mathsf{G}^{-1}, \tag{2.9}$$

where $\mathsf{G}$ is a permutation matrix that reorders the states into pairs. The set of nonzero eigenvalues is identical for the $A$ and $B$ subspaces, but if these are of different dimensions ($N_a \neq N_b$) then the larger set will include also null eigenvalues associated with decoupled states, also known as multilevel dark states [9,11,14]. For example, in the five-state $M$ system of Fig. 1, the dimensions of the subspace $A$ is $N_a = 3 = N_>$, and the diagonalization (of basis states 1, 2, and 3) produces the results

$$\sqrt{\mathsf{A}\mathsf{V}\mathsf{V}^\dagger\mathsf{A}^\dagger} = \tfrac{1}{2}\{|\Omega^{(1)}|, |\Omega^{(2)}|, 0\}. \tag{2.10}$$

In the $B$ subspace of dimension $N_b = 2 = N_<$ and involving basis states 4 and 5, the null eigenvalue is missing,

$$\sqrt{\mathsf{B}\mathsf{V}^\dagger\mathsf{V}\mathsf{B}^\dagger} = \tfrac{1}{2}\{|\Omega^{(1)}|, |\Omega^{(2)}|\}. \tag{2.11}$$

The phases of the eigenvalues (the Rabi frequencies $\Omega^{(m)}$) are obtained only with the evaluation of $\overline{\mathsf{W}} = \mathsf{U}\mathsf{W}\mathsf{U}^\dagger$.

With appropriate reorganization of the MS states, $\overline{C}_n^{(m)} \equiv \overline{C}_{2m+n-2}$, one has the equation

$$\frac{d}{dt}\begin{bmatrix} \overline{C}_1^{(m)} \\ \overline{C}_2^{(m)} \end{bmatrix} = -i \begin{bmatrix} \Delta_a & \frac{1}{2}\Omega^{(m)} \\ \frac{1}{2}\Omega^{(m)} & \Delta_b \end{bmatrix} \begin{bmatrix} \overline{C}_1^{(m)} \\ \overline{C}_2^{(m)} \end{bmatrix}. \tag{2.12}$$

The detunings in this last formula are unchanged from the original problem. The Rabi frequency $\Omega^{(m)}$ for the $m$th two-state system is obtained from square roots of the eigenvalues of the matrix $\mathsf{V}\mathsf{V}^\dagger$ or $\mathsf{V}^\dagger\mathsf{V}$, as explained above. Because, by assumption, all of the original Rabi frequencies share a common time dependence, the eigenvalue frequencies $\Omega^{(m)}$ share this same time dependence.

Although the original Rabi frequencies may have very simple symmetries, involving repetition of common values (they may be proportional to Clebsch-Gordon coefficients, for example), the eigenvalue Rabi frequencies $\Omega^{(m)}$ of the two-state systems are generally not degenerate: they are all different.

The Morris-Shore transformation is quite general (given the constraint of only two distinct detunings). It can be used, for example, with various "bent" linkages ($\Lambda$, $V$, $M$, and $W$), with various many-to-one connections ($\Lambda$, the tripod, . . . ), as well as with complicated hyperfine interactions. It can also be used with a resonant $N$-state ladder of excitation: alternating states of the chain are placed into the $A$ and $B$ set.

## III. ANALYTIC SOLUTIONS IN MULTISTATE SYSTEMS

### A. Time evolution in the original and MS bases

The original probability amplitudes $C_n(t)$ are obtained from the MS amplitudes $\overline{C}_j(t)$ by means of the time-independent unitary transformation,

$$C_n(t) = \sum_j U^\dagger_{nj} \overline{C}_j(t). \tag{3.1}$$

To obtain probabilities $P_n(t)$ for the physical basis states one therefore requires a coherent (but time-independent) superposition of the MS states,

$$P_n(t) = |C_n(t)|^2 = \sum_{ij} U^\dagger_{nj} U^T_{ni} \overline{C}_j(t) \overline{C}^*_i(t). \tag{3.2}$$

The time development of each MS amplitude $\overline{C}^{(m)}_n(t)$ can be obtained by solving the two coupled equations (2.12). Each solution, at a fixed time $t$, is expressible in terms of the initial amplitudes at time $t=t_0$ by a unitary transformation

$$\overline{C}^{(m)}_n(t) = \sum_{k=1,2} \overline{S}^{(m)}_{nk}(t,t_0) \overline{C}^{(m)}_k(t_0) \quad (m=1, \ldots, N_<). \tag{3.3}$$

The original probability amplitudes are then obtained as

$$C_n(t) = \sum_k S_{nk}(t,t_0) C_k(t_0), \tag{3.4}$$

where the transition matrix is

$$\mathsf{S}(t,t_0) = \mathsf{U}^\dagger \overline{\mathsf{S}}(t,t_0) \mathsf{U}, \tag{3.5a}$$

$$\overline{\mathsf{S}}(t,t_0) = \left( \bigoplus_{m=1}^{N_<} \overline{\mathsf{S}}^{(m)}(t,t_0) \right) \oplus \mathsf{I}. \tag{3.5b}$$

Here the transition matrix is expressed as a direct sum of independent two-state transition matrices $\overline{\mathsf{S}}^{(m)}(t,t_0)$ together with $\mathsf{I}$, a unit matrix of dimension $N_> - N_<$.

Although the original Hamiltonian has been replaced by a set of independent two-state systems, the complexity of the original problem returns when one evaluates the original populations,

$$P_n(t) = |C_n(t)|^2 = \left| \sum_k S_{nk}(t) C_k(t_0) \right|^2. \tag{3.6}$$

From this expression it is clear that interference between the independent two-state systems can contribute to the observable population changes.

Even when the original physical system starts in a single state, this initial condition will appear as a superposition of the MS amplitudes. For example, let the single initial physical state be labeled $n=1$. Then the initial MS amplitudes are $\overline{C}_j(t_0) = U_{j1}$ and Eq. (3.6) reads

$$P_n(t) = |S_{n1}(t,t_0)|^2. \tag{3.7}$$

Interference still occurs between the separate two-state histories. Whether this is constructive or destructive (or something intermediate) depends on the elements of the $\mathsf{S}$ matrix, which in turn depend not only on the time interval $(t,t_0)$ but also on the original Rabi frequencies, detunings and linkage pattern.

### B. Analytic solutions

The procedure outlined above, for reducing a class of degenerate multilevel systems into a set of independent two-state systems, simplifies considerably the problem of finding analytic solutions of the Schrödinger equation (2.1) with the Hamiltonian $\hbar \mathsf{W}$ defined by Eq. (2.2). In the MS basis the Hamiltonian $\hbar \overline{\mathsf{W}}$ has block-diagonal form with one- and two-dimensional submatrices. The $2\times 2$ blocks describe two-state systems undergoing time evolution, while the remaining elements are associated with decoupled states.

Numerous analytically soluble two-state models have been described. We can use any of these with Eq. (2.12). However, each $2\times 2$ block $\mathsf{w}^{(m)}$ must have the same pair of detunings (perhaps time dependent) and the same time dependence (if any) for the Rabi frequencies.

The algebra is simplified when the detunings are symmetrically distributed,

$$-\Delta_a = \Delta_b = \frac{\Delta}{2}. \tag{3.8}$$

In this case the $2\times 2$ transfer matrix of Eq. (3.3) can be parametrized as

$$\overline{\mathsf{S}}^{(n)}(t) = \begin{bmatrix} \alpha^{(n)} & -(\beta^{(n)})^* \\ \beta^{(n)} & (\alpha^{(n)})^* \end{bmatrix}. \tag{3.9}$$

The transition probability between the lower and upper state of a pair of MS basis states is $|\beta^{(n)}|^2$, and the probability for no transition is $|\alpha^{(n)}|^2 = 1 - |\beta^{(n)}|^2$. The Appendix provides a number of examples of these matrices for several analytically soluble two-state systems.

The decoupled states will also undergo a time evolution, acquiring a phase factor

$$\kappa = \exp\left[ \frac{i}{2} \int_{t_0}^t \Delta(t) dt \right]. \tag{3.10}$$

Analytic solutions can reveal various properties of the population dynamics of systems with complicated linkage patterns. Such solutions allow one to estimate the maximal population any particular state can obtain for chosen initial conditions. In particular, one can derive conditions for complete population transfer between any two states, such as generalized $\pi$ pulses.

## IV. APPLICATIONS TO DEGENERATE FIVE-STATE CHAINS

### A. Example linkages

The properties of the MS transformation, and the novelty, can be illustrated by reconsidering the system mentioned by Morris and Shore [7], who considered only time-independent Rabi frequencies. We consider transitions produced between angular momentum states (specified by $J$ and $M$) produced by elliptically polarized light, and we take the quantization axis to be the laser propagation direction, i.e., we describe the polarization by means of two helicity states. The selection rule for electric-dipole interaction (assumed) then limits transitions to those for which $\Delta M=\pm 1$. We consider two cases: the $M$-linkage pattern (see Fig. 1) found for $J_a=2$ to $J_b=1$ or $J_b=2$, and the $W$ linkage (see Fig. 2) found for $J_a=1$ or $J_a=2$ to $J_b=2$. For each of these systems the MS transformation produces a Hamiltonian that consists of two separate $2\times 2$ matrices and one decoupled state.

### B. Elliptical polarization

The couplings described by Figs. 1 and 2 can be produced by a single elliptically polarized laser whose electric field has the form [15]

$$\mathbf{E}(t)=\tfrac{1}{2}[\mathbf{e}_{+1}\mathcal{E}_{+1}(t)e^{-i\omega t}+\mathbf{e}_{-1}\mathcal{E}_{-1}(t)e^{-i\omega t}+\text{c.c.}]. \tag{4.1}$$

Here $\mathbf{e}_q$ is a unit vector, of helicity $q=\pm 1$, as is appropriate for the expression of elliptical polarization as a combination of circular polarizations $\sigma^+$ and $\sigma^-$. The needed matrix elements of the electric-dipole interaction can then be written, with the aid of the Wigner-Eckart theorem (cf. Ref. [4]) as

$$\langle J_bM_b|\mathbf{d}\cdot\mathbf{E}(t)e^{i\omega t}|J_aM_a\rangle=\frac{1}{2}\hbar\Omega_q\xi_{M_a}^{M_b}e^{i(\omega t-\phi_q)}, \tag{4.2}$$

where the constant parameters $\phi_+$ and $\phi_-$ are the phases of the $\sigma^+$ and $\sigma^-$ fields. The dependence on magnetic quantum numbers occurs through a Clebsch-Gordon coefficient, which we incorporate into the (constant) relative coupling strength

$$\xi_{M_a}^{M_b}=(J_aM_a,1q|J_bM_b)/\sqrt{2J_a+1}. \tag{4.3}$$

The two real-valued quantities $\Omega_\pm\equiv\Omega_{\pm q}$ are time dependent (pulse-shaped) Rabi frequency "units" for the couplings induced by the two independent polarizations. These must share the same time dependence but may have different peak values.

The ellipticity of the laser pulse, defined by

$$\varepsilon=\frac{\Omega_+^2-\Omega_-^2}{\Omega_+^2+\Omega_-^2}=\frac{\mathcal{E}_+^2-\mathcal{E}_-^2}{\mathcal{E}_+^2+\mathcal{E}_-^2}=\frac{I_+-I_-}{I_++I_-}, \tag{4.4}$$

where $I_\pm$ are the corresponding intensities and $\mathcal{E}_\pm\equiv\mathcal{E}_{\pm 1}$, provides a useful means of parametrizing the polarization. Ellipticity $\varepsilon=\pm 1$ corresponds to $\sigma^\pm$ polarization and $\varepsilon=0$ to linear polarization. The Rabi frequencies $\Omega_+$ and $\Omega_-$ are conveniently parametrized in terms of $\varepsilon$ and $\Omega=\sqrt{\Omega_+^2+\Omega_-^2}$ as $\Omega_\pm=\Omega\sqrt{\frac{1}{2}(1\pm\varepsilon)}$.

### C. The *M* linkage

In the $M$ linkage the lower level has $J_a=2$ and the upper has $J_b=1$ or $J_b=2$. We label the physical states by the magnetic quantum number $M$. States $\{-2,0,+2\}$ are part of the lower-level manifold, while states $\{-1,+1\}$ form the upper-level manifold, see Fig. 1. The $3\times 2$ interaction matrix $\mathsf{V}$ has the elements

$$\mathsf{V}=\frac{1}{2}\begin{bmatrix}\xi_{-2}^{-1}\Omega_+e^{i\phi_+} & 0\\ \xi_0^{-1}\Omega_-e^{-i\phi_-} & \xi_0^1\Omega_+e^{i\phi_+}\\ 0 & \xi_2^1\Omega_-e^{-i\phi_-}\end{bmatrix}. \tag{4.5}$$

For $J_a=2\leftrightarrow J_b=1$ the Clebsch-Gordon coefficients $\xi_{M_a}^{M_b}$ are given by $\xi_{-2}^{-1}=\xi_2^1=\sqrt{\frac{3}{5}}$, $\xi_0^{-1}=\xi_0^1=\sqrt{\frac{1}{10}}$, while for $J_a=2\leftrightarrow J_b=2$ they are $\xi_{-2}^{-1}=-\xi_2^1=-\sqrt{\frac{1}{3}}$, $\xi_0^{-1}=-\xi_0^1=\sqrt{\frac{1}{2}}$.

### D. The *W* linkage

In the $W$ linkage the lower level has $J_a=1$ or $J_a=2$ and the upper has $J_b=2$. As in the case of the $M$ linkage, we label the physical states by the magnetic quantum number $M$. States $\{-1,+1\}$ are part of the lower-level manifold, while states $\{-2,0,+2\}$ form the upper-level manifold, see Fig. 2. The interaction matrix is that of Eq. (4.5), but with the states relabeled, i.e., the indices $M_a$ and $M_b$ are interchanged.

### E. The MS transformation for the *M* and *W* linkages

The MS transformation replaces the usual atomic basis states by a set of new MS basis states. In our case, the Rabi frequencies of the Hamiltonian in the new MS basis are obtained from the square roots of the eigenvalues of the matrices

$$\mathsf{V}\mathsf{V}^\dagger=\frac{1}{4}\begin{bmatrix}(\xi_{-2}^{-1})^2\Omega_+^2 & \xi_{-2}^{-1}\xi_0^{-1}\Omega_+\Omega_-e^{i\phi} & 0\\ \xi_{-2}^{-1}\xi_0^{-1}\Omega_+\Omega_-e^{-i\phi} & (\xi_0^{-1})^2\Omega_-^2+(\xi_0^1)^2\Omega_+^2 & \xi_0^1\xi_2^1\Omega_+\Omega_-e^{i\phi}\\ 0 & \xi_0^1\xi_2^1\Omega_+\Omega_-e^{-i\phi} & (\xi_2^1)^2\Omega_-^2\end{bmatrix}, \tag{4.6a}$$

$$\mathsf{V}^\dagger\mathsf{V}=\frac{1}{4}\begin{bmatrix}(\xi_{-2}^{-1})^2\Omega_+^2+(\xi_0^{-1})^2\Omega_-^2 & \xi_0^{-1}\xi_0^1\Omega_+\Omega_-e^{i\phi}\\ \xi_0^{-1}\xi_0^1\Omega_+\Omega_-e^{-i\phi} & (\xi_0^1)^2\Omega_+^2+(\xi_2^1)^2\Omega_-^2\end{bmatrix} \tag{4.6b}$$

with $\phi=\phi_++\phi_-$.

The three eigenvalues of $\mathsf{V}\mathsf{V}^\dagger$ provide the squares of MS Rabi frequencies $\Omega^{(n)}$. One of these is zero, $\Omega^{(0)}=0$, while the other, nonzero ones are ($n=1,2$)

TABLE I. The coefficients for the MS transformation for $J_a=2\leftrightarrow J_b=1$ (first column) and $J_a=2\leftrightarrow J_b=2$ (second column) cases [9]. Note that $c_-^{(n)}(-\varepsilon)=-c_+^{(n)}(\varepsilon)$ and $c_0^{(n)}(-\varepsilon)=-c_0^{(n)}(\varepsilon)$ for $n=1,2$.

| | $J_a=2\leftrightarrow J_b=1$ | $J_a=2\leftrightarrow J_b=2$ |
|---|---|---|
| $d_-(\varepsilon)$ | $\nu_d(1-\varepsilon)$ | $\nu_d\sqrt{3}(1-\varepsilon)$ |
| $d_0(\varepsilon)$ | $-\nu_d\sqrt{6(1-\varepsilon^2)}$ | $\nu_d\sqrt{2(1-\varepsilon^2)}$ |
| $d_+(\varepsilon)$ | $\nu_d(1+\varepsilon)$ | $\nu_d\sqrt{3}(1+\varepsilon)$ |
| $[\nu_d(\varepsilon)]^{-2}$ | $4(2-\varepsilon^2)$ | $4(2+\varepsilon^2)$ |
| $c_-^{(1)}(\varepsilon)$ | $-\frac{1}{2}\nu_c^{(1)}(1+\varepsilon)(1-6\varepsilon-\sqrt{1+24\varepsilon^2})$ | $\frac{1}{2}\nu_c^{(1)}(1+\varepsilon)(3-2\varepsilon-\sqrt{9-8\varepsilon^2})$ |
| $c_0^{(1)}(\varepsilon)$ | $\nu_c^{(1)}\varepsilon\sqrt{6(1-\varepsilon^2)}$ | $\nu_c^{(1)}\varepsilon\sqrt{6(1-\varepsilon^2)}$ |
| $c_+^{(1)}(\varepsilon)$ | $\frac{1}{2}\nu_c^{(1)}(1-\varepsilon)(1+6\varepsilon-\sqrt{1+24\varepsilon^2})$ | $-\frac{1}{2}\nu_c^{(1)}(1-\varepsilon)(3+2\varepsilon-\sqrt{9-8\varepsilon^2})$ |
| $[\nu_c^{(1)}(\varepsilon)]^{-2}$ | $\sqrt{1+24\varepsilon^2}[(1+\varepsilon^2)\sqrt{1+24\varepsilon^2}+11\varepsilon^2-1]$ | $\sqrt{9-8\varepsilon^2}[(1+\varepsilon^2)\sqrt{9-8\varepsilon^2}+\varepsilon^2-3]$ |
| $c_-^{(2)}(\varepsilon)$ | $-\frac{1}{2}\nu_c^{(1)}(1+\varepsilon)(1-6\varepsilon+\sqrt{1+24\varepsilon^2})$ | $\frac{1}{2}\nu_c^{(2)}(1+\varepsilon)(3-2\varepsilon+\sqrt{9-8\varepsilon^2})$ |
| $c_0^{(2)}(\varepsilon)$ | $\nu_c^{(2)}\varepsilon\sqrt{6(1-\varepsilon^2)}$ | $\nu_c^{(1)}\varepsilon\sqrt{6(1-\varepsilon^2)}$ |
| $c_+^{(2)}(\varepsilon)$ | $\frac{1}{2}\nu_c^{(2)}(1-\varepsilon)(1+6\varepsilon+\sqrt{1+24\varepsilon^2})$ | $-\frac{1}{2}\nu_c^{(2)}(1-\varepsilon)(3+2\varepsilon+\sqrt{9-8\varepsilon^2})$ |
| $[\nu_c^{(2)}(\varepsilon)]^{-2}$ | $\sqrt{1+24\varepsilon^2}[(1+\varepsilon^2)\sqrt{1+24\varepsilon^2}-11\varepsilon^2+1]$ | $\sqrt{9-8\varepsilon^2}[(1+\varepsilon^2)\sqrt{9-8\varepsilon^2}-\varepsilon^2+3]$ |
| $e_-^{(1)}(\varepsilon)$ | $\nu_e(\sqrt{1+24\varepsilon^2}+5\varepsilon)^{1/2}$ | $\nu_e(\sqrt{9-8\varepsilon^2}-\varepsilon)^{1/2}$ |
| $e_+^{(1)}(\varepsilon)$ | $\nu_e(\sqrt{1+24\varepsilon^2}-5\varepsilon)^{1/2}$ | $\nu_e(\sqrt{9-8\varepsilon^2}+\varepsilon)^{1/2}$ |
| $e_-^{(2)}(\varepsilon)$ | $\nu_e(\sqrt{1+24\varepsilon^2}-5\varepsilon)^{1/2}$ | $\nu_e(\sqrt{9-8\varepsilon^2}+\varepsilon)^{1/2}$ |
| $e_+^{(2)}(\varepsilon)$ | $-\nu_e(\sqrt{1+24\varepsilon^2}+5\varepsilon)^{1/2}$ | $-\nu_e(\sqrt{9-8\varepsilon^2}-\varepsilon)^{1/2}$ |
| $[\nu_e(\varepsilon)]^{-2}$ | $2\sqrt{1+24\varepsilon^2}$ | $2\sqrt{9-8\varepsilon^2}$ |

$$\Omega^{(n)}(t)=\sqrt{\frac{1}{20}[7-(-1)^n\sqrt{1+24\varepsilon^2}]}\Omega(t), \tag{4.7a}$$

$$\Omega^{(n)}(t)=\sqrt{\frac{1}{12}[5-(-1)^n\sqrt{9-8\varepsilon^2}]}\Omega(t), \tag{4.7b}$$

for $J_a=2\leftrightarrow J_b=1$ and $J_a=2\leftrightarrow J_b=2$, respectively. Here $\Omega^{(1)}$ corresponds to the larger eigenvalue, with a plus sign in Eqs. (4.7), and $\Omega^{(2)}$ to the smaller eigenvalue, with a minus sign. The eigenvectors of $\mathsf{V}\mathsf{V}^\dagger$ are superpositions of ground states: two MS coupled (bright) states $\varphi_c^{(1)}(\varepsilon)$ and $\varphi_c^{(2)}(\varepsilon)$ and a decoupled (dark) state $\varphi_d(\varepsilon)$, given by the constructions

$$\varphi_c^{(1)}(\varepsilon)=c_-^{(1)}(\varepsilon)\psi_{-2}+c_0^{(1)}(\varepsilon)e^{-i\phi}\psi_0+c_+^{(1)}(\varepsilon)e^{-2i\phi}\psi_{+2}, \tag{4.8a}$$

$$\varphi_c^{(2)}(\varepsilon)=c_-^{(2)}(\varepsilon)\psi_{-2}+c_0^{(2)}(\varepsilon)e^{-i\phi}\psi_0+c_+^{(2)}(\varepsilon)e^{-2i\phi}\psi_{+2}, \tag{4.8b}$$

$$\varphi_d(\varepsilon)=d_-(\varepsilon)\psi_{-2}+d_0(\varepsilon)e^{-i\phi}\psi_0+d_+(\varepsilon)e^{-2i\phi}\psi_{+2}. \tag{4.8c}$$

The eigenvectors of $\mathsf{V}^\dagger\mathsf{V}$ are superpositions of excited states only,

$$\varphi_e^{(1)}(\varepsilon)=e_-^{(1)}(\varepsilon)\psi_{-1}+e_+^{(1)}(\varepsilon)e^{-i\phi}\psi_1, \tag{4.9a}$$

$$\varphi_e^{(2)}(\varepsilon)=e_-^{(2)}(\varepsilon)\psi_{-1}+e_+^{(2)}(\varepsilon)e^{-i\phi}\psi_1, \tag{4.9b}$$

and the eigenvalues of $\mathsf{V}^\dagger\mathsf{V}$ are given by Eqs. (4.7). The parameters of these new basis states are given in Table I. Their values for the special cases $\varepsilon=0,\pm1$ are given in Table II.

We emphasize here that the five MS states, Eqs. (4.8) and (4.9)—the decoupled state, the two coupled states, and the two excited states—are entirely determined by the laser polarization parameters $\varepsilon$ and $\phi$.

The transformation matrix between the original amplitudes and the MS amplitudes is constructed from the normalized eigenstates of the Hamiltonian. It is given by Eq. (2.9), where

TABLE II. The coefficients for the MS transformation for $J_a=2\leftrightarrow J_b=1$ and $J_a=2\leftrightarrow J_b=2$ for the special values of the ellipticity $\varepsilon=0,\pm1$.

| | $\varepsilon$ | $d_-$ | $d_0$ | $d_+$ | $c_-^{(1)}$ | $c_0^{(1)}$ | $c_+^{(1)}$ | $c_-^{(2)}$ | $c_0^{(2)}$ | $c_+^{(2)}$ | $e_-^{(1)}$ | $e_+^{(1)}$ | $e_-^{(2)}$ | $e_+^{(2)}$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 0 | $\sqrt{\frac{1}{8}}$ | $-\sqrt{\frac{3}{4}}$ | $\sqrt{\frac{1}{8}}$ | $\sqrt{\frac{3}{8}}$ | $\frac{1}{2}$ | $\sqrt{\frac{3}{8}}$ | $-\sqrt{\frac{1}{2}}$ | 0 | $\sqrt{\frac{1}{2}}$ | $\sqrt{\frac{1}{2}}$ | $\sqrt{\frac{1}{2}}$ | $\sqrt{\frac{1}{2}}$ | $-\sqrt{\frac{1}{2}}$ |
| $J_a=2\leftrightarrow J_b=1$ | $-1$ | 1 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 1 | 0 | 1 | 1 | 0 |
| | 1 | 0 | 0 | 1 | 1 | 0 | 0 | 1 | 0 | 0 | 1 | 0 | 0 | 1 |
| | 0 | $\sqrt{\frac{3}{8}}$ | $\frac{1}{2}$ | $\sqrt{\frac{3}{8}}$ | $\sqrt{\frac{1}{8}}$ | $-\sqrt{\frac{3}{4}}$ | $-\sqrt{\frac{1}{8}}$ | $\sqrt{\frac{1}{2}}$ | 0 | $-\sqrt{\frac{1}{2}}$ | $\sqrt{\frac{1}{2}}$ | $\sqrt{\frac{1}{2}}$ | $\sqrt{\frac{1}{2}}$ | $-\sqrt{\frac{1}{2}}$ |
| $J_a=2\leftrightarrow J_b=2$ | $-1$ | 1 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 1 | 1 | 0 | 0 | 1 |
| | 1 | 0 | 0 | 1 | 1 | 0 | 0 | 1 | 0 | 0 | 0 | 1 | 1 | 0 |

TABLE III. Final populations for the $M$ system for initial state $\psi_{-2}$ and linear polarization ($\varepsilon=0$).

| | $J_a=2\leftrightarrow J_b=1$ | $J_a=2\leftrightarrow J_b=2$ |
|---|---|---|
| $P_{-2}$ | $\lvert\frac{1}{8}\kappa+\frac{3}{8}\alpha^{(1)}+\frac{1}{2}\alpha^{(2)}\rvert^2$ | $\lvert\frac{3}{8}\kappa+\frac{1}{8}\alpha^{(1)}+\frac{1}{2}\alpha^{(2)}\rvert^2$ |
| $P_0$ | $\frac{3}{32}\lvert\kappa-\alpha^{(1)}\rvert^2$ | $\frac{3}{32}\lvert\kappa-\alpha^{(1)}\rvert^2$ |
| $P_2$ | $\lvert\frac{1}{8}\kappa+\frac{3}{8}\alpha^{(1)}-\frac{1}{2}\alpha^{(2)}\rvert^2$ | $\lvert\frac{3}{8}\kappa+\frac{1}{8}\alpha^{(1)}-\frac{1}{2}\alpha^{(2)}\rvert^2$ |
| $P_{-1}$ | $\lvert\sqrt{\frac{3}{16}}\beta^{(2)}-\frac{1}{2}\beta^{(2)}\rvert^2$ | $\lvert\frac{1}{4}\beta^{(1)}-\frac{1}{2}\beta^{(2)}\rvert^2$ |
| $P_1$ | $\lvert\sqrt{\frac{3}{16}}\beta^{(2)}+\frac{1}{2}\beta^{(2)}\rvert^2$ | $\lvert\frac{1}{4}\beta^{(1)}+\frac{1}{2}\beta^{(2)}\rvert^2$ |

$$\mathsf{G}=\begin{bmatrix}1&0&0&0&0\\0&1&0&0&0\\0&0&0&1&0\\0&0&1&0&0\\0&0&0&0&1\end{bmatrix}, \tag{4.10a}$$

$$\begin{bmatrix}\mathsf{A}&0\\0&\mathsf{B}\end{bmatrix}=\begin{bmatrix}d_-&d_0e^{-i\phi}&d_+e^{-2i\phi}&0&0\\c_-^{(1)}&c_0^{(1)}e^{-i\phi}&c_+^{(1)}e^{-2i\phi}&0&0\\c_-^{(2)}&c_0^{(2)}e^{-i\phi}&c_+^{(2)}e^{-2i\phi}&0&0\\0&0&0&e_-^{(1)}&e_+^{(1)}e^{-i\phi}\\0&0&0&e_-^{(2)}&e_+^{(2)}e^{-i\phi}\end{bmatrix}. \tag{4.10b}$$

## V. THE *M* SYSTEM

### A. Populations in the general case

If the system is initially in state $\psi_{-2}$, i.e., $\mathbf{C}(-\infty)=[1,0,0,0,0]^T$, the final populations are given by the squared moduli of the elements in the first column of the evolution matrix $\mathsf{U}$, $P_M=|\mathsf{U}_{1M}|^2$,

$$P_{-2}=|\kappa d_-^2+\alpha^{(1)}c_-^{(1)2}+\alpha^{(2)}c_-^{(2)2}|^2, \tag{5.1a}$$

$$P_0=|\kappa d_-d_0+\alpha^{(1)}c_-^{(1)}c_0^{(1)}+\alpha^{(2)}c_-^{(2)}c_0^{(2)}|^2, \tag{5.1b}$$

$$P_2=|\kappa d_-d_++\alpha^{(1)}c_-^{(1)}c_+^{(1)}+\alpha^{(2)}c_-^{(2)}c_+^{(2)}|^2, \tag{5.1c}$$

$$P_{-1}=|\beta^{(1)*}c_-^{(1)}e_-^{(1)}+\beta^{(2)*}c_-^{(2)}e_-^{(2)}|^2, \tag{5.1d}$$

$$P_1=|\beta^{(1)*}c_-^{(1)}e_+^{(1)}+\beta^{(2)*}c_-^{(2)}e_+^{(2)}|^2. \tag{5.1e}$$

For linear polarization ($\varepsilon=0$) we summarize the values of these populations in Table III. Equations (5.1) and the analytic formulas for the two-state evolution-matrix elements $\alpha^{(n)}$ and $\beta^{(n)}$ provide several analytic solutions for the $M$ system.

The coefficients $(d_-,d_0,d_+)$ of the decoupled state $\varphi_d$ appear in the populations of the ground sublevels. Their contribution depends only on the ellipticity $\varepsilon$ of the laser.

Because the decoupled state does not interact with the laser pulse, its population,

$$P_d=\frac{(1-\varepsilon)^2}{4(2-\varepsilon^2)}\quad(J_a=2\leftrightarrow J_b=1), \tag{5.2a}$$

$$P_d=\frac{3(1-\varepsilon)^2}{4(2+\varepsilon^2)}\quad(J_a=2\leftrightarrow J_b=2), \tag{5.2b}$$

is conserved. This population is trapped and excluded from the excitation process. For example, for $\varepsilon=0$, the trapped population is $P_d=\frac{1}{8}$ and $P_d=\frac{3}{8}$, in the two cases $2\leftrightarrow1$ and $2\leftrightarrow2$, respectively.

### B. All population in the ground sublevels

In this section we present the conditions needed to ensure that, following a pulse, all population remains in the low-lying initially populated sublevels, with none in the excited sublevels.

#### *1. General case*

All population will be confined to the ground sublevels if the excited-level populations vanish, $P_{-1}=P_1=0$. Because states $\psi_{-1}$ and $\psi_1$ participate only in the MS states $\varphi_e^{(1)}$ and $\varphi_e^{(2)}$, this condition requires no transition in the MS basis,

$$\beta^{(1)}=\beta^{(2)}=0. \tag{5.3}$$

This means that the transition probabilities in both of the MS two-state systems should vanish simultaneously, i.e., the evolution matrix $\bar{\mathsf{S}}$ in Eq. (3.5b) should be diagonal. In the following sections we present several analytically solvable models for two-state systems and find the parameters that satisfy Eq. (5.3).

#### *2. Exact resonance*

On exact resonance, the elements of the transfer matrix (3.9) take a particularly simple form, Eq. (A4), for arbitrary pulse shape. In this case the phase factor of the decoupled state $\kappa$, Eq. (3.10), is equal to unity. In order to satisfy conditions (5.3), the two pulse areas in the MS two-state systems should be even integer multiples of $\pi$,

$$\mathcal{A}^{(n)}=k^{(n)}\pi\quad(n=1,2), \tag{5.4}$$

where the pulse area is defined by Eq. (A3) and

$$k^{(n)}=2m^{(n)}, \tag{5.5}$$

where $m^{(n)}$ is an integer. Explicitly, for $J_a=2\leftrightarrow J_b=1$ and $J_a=2\leftrightarrow J_b=2$ we have, respectively,

$$\mathcal{A}^{(n)}=\sqrt{\tfrac{1}{20}[7-(-1)^n\sqrt{1+24\varepsilon^2}]}\mathcal{A}=k^{(n)}\pi, \tag{5.6a}$$

$$\mathcal{A}^{(n)}=\sqrt{\tfrac{1}{12}[5-(-1)^n\sqrt{9-8\varepsilon^2}]}\mathcal{A}=k^{(n)}\pi. \tag{5.6b}$$

These conditions can be satisfied for an infinite number of pairs of pulse areas and ellipticities $(\mathcal{A},\varepsilon)$. The solution for $\mathcal{A}$ and $\varepsilon$ in the case $J_a=2\leftrightarrow J_b=1$ reads

$$\mathcal{A}=\pi\sqrt{\tfrac{10}{7}(k^{(1)2}+k^{(2)2})}, \tag{5.7a}$$

$$\varepsilon=\pm\sqrt{\frac{(4k^{(1)2}-3k^{(2)2})(3k^{(1)2}-4k^{(2)2})}{6(k^{(1)2}+k^{(2)2})^2}}, \quad (5.7b)$$

and for $J_a=2\leftrightarrow J_b=2$ it is

$$\mathcal{A}=\pi\sqrt{\tfrac{6}{5}(k^{(1)2}+k^{(2)2})}, \quad (5.8a)$$

$$\varepsilon=\pm\sqrt{\frac{(4k^{(2)2}-k^{(1)2})(4k^{(1)2}-k^{(2)2})}{2(k^{(1)2}+k^{(2)2})^2}}. \quad (5.8b)$$

Only such pairs $(k^{(1)},k^{(2)})$ apply, for which $\varepsilon$ is real and $|\varepsilon|\leqslant 1$; these conditions require

$$\sqrt{\frac{1}{6}}\leqslant\frac{k^{(2)}}{k^{(1)}}<1 \quad (J_a=2\leftrightarrow J_b=1), \quad (5.9a)$$

$$\frac{1}{2}\leqslant\frac{k^{(2)}}{k^{(1)}}\leqslant\sqrt{\frac{2}{3}} \quad (J_a=2\leftrightarrow J_b=2). \quad (5.9b)$$

Obviously, there are infinitely many pairs $(k^{(1)},k^{(2)})$, which satisfy these conditions.

#### 3. Off-resonant rectangular pulse

For a rectangular pulse with a constant detuning, we conclude from Eqs. (A7) that conditions (5.3) require the fulfillment of the following equations:

$$\sqrt{\Omega^{(1)2}+\Delta^2}T=2m^{(1)}\pi, \quad (5.10a)$$

$$\sqrt{\Omega^{(2)2}+\Delta^2}T=2m^{(2)}\pi, \quad (5.10b)$$

where $m^{(n)}$ are integers, and $\Omega^{(1)}$ and $\Omega^{(2)}$ are given by Eqs. (4.7). The solution for $\Omega$ and $\Delta$ in the case $J_a=2\leftrightarrow J_b=1$ reads

$$\Omega=\frac{2\pi}{T}\sqrt{\frac{10}{\sqrt{1+24\varepsilon^2}}(m^{(1)2}-m^{(2)2})}, \quad (5.11a)$$

$$\Delta=\frac{2\pi}{T}\sqrt{\frac{m^{(1)2}+m^{(2)2}}{2}-\frac{7(m^{(1)2}-m^{(2)2})}{2\sqrt{1+24\varepsilon^2}}}, \quad (5.11b)$$

and for $J_a=2\leftrightarrow J_b=2$ it is

$$\Omega=\frac{2\pi}{T}\sqrt{\frac{6}{\sqrt{9-8\varepsilon^2}}(m^{(1)2}-m^{(2)2})}, \quad (5.12a)$$

$$\Delta=\frac{2\pi}{T}\sqrt{\frac{m^{(1)2}+m^{(2)2}}{2}-\frac{5(m^{(1)2}-m^{(2)2})}{2\sqrt{9-8\varepsilon^2}}}. \quad (5.12b)$$

In the above equations the polarization $\varepsilon$ is a free parameter. We use this leeway below for finding cases of complete population transfer from $\psi_{-2}$ to $\psi_2$.

#### 4. Other analytic models with detuning

We shall now discuss the possibility of satisfying the conditions (5.3) for the other analytically soluble nonresonant models. First of all, conditions (5.3) are satisfied automatically in the trivial case of no interaction (all $\Omega$'s zero), which is of no interest; we therefore assume that the Rabi frequencies are nonzero.

For the Landau-Zener model (Appendix, Sec. 3), conditions (5.3) cannot be satisfied, cf. Eq. (A10).

For the Demkov-Kunike model (Appendix, Sec. 4), it follows from Eq. (A13) that conditions (5.3) can be satisfied only for constant detuning $(B=0)$. Hence these conditions cannot be satisfied for the Allen-Eberly-Hioe and Bambini-Berman models (Appendix, Secs. 4 c and 4 d), which involve chirped detuning $(B\neq 0)$; for such detuning some population is always left in the excited states of the $M$ system. Hence, chirped pulses are not suitable when the goal is to confine the population dynamics in the ground sublevels.

For the Rosen-Zener model (Appendix, Sec. 4 b), conditions (5.3) are satisfied for the same pulse areas (note that $\mathcal{A}=\pi\Omega_0 T$ for the hyperbolic-secant pulse) as for exact resonance, cf. Eqs. (A5) and (A15); this is a peculiarity of this model only.

### C. Complete population transfer between the ground sublevels $\psi_{-2}\rightarrow\psi_2$

Redistribution of population amongst magnetic sublevels, thereby producing orientation or alignment, is commonly accomplished by means of optical pumping [16]. Here we consider the task of transferring population, via coherent processes alone, between sublevels.

#### 1. General case

An important special case of population residing in the ground sublevels is the complete population transfer (CPT) from state $\psi_{-2}$ to state $\psi_2$. In this case, in addition to the requirement $P_{-1}=P_1=0$, which led us to conditions (5.3), we must also have $P_{-2}=P_0=0$, i.e. [cf. Eqs. (5.1a) and (5.1b)],

$$\kappa d_-^2+\alpha^{(1)}c_-^{(1)2}+\alpha^{(2)}c_-^{(2)2}=0, \quad (5.13a)$$

$$\kappa d_-d_0+\alpha^{(1)}c_-^{(1)}c_0^{(1)}+\alpha^{(2)}c_-^{(2)}c_0^{(2)}=0. \quad (5.13b)$$

The solution for $\alpha^{(1)}$ and $\alpha^{(2)}$ reads

$$\alpha^{(1)}=\frac{(\varepsilon-1)(5\varepsilon-\sqrt{1+24\varepsilon^2})}{(\varepsilon+1)^2}\kappa, \quad (5.14a)$$

$$\alpha^{(2)}=\frac{(\varepsilon-1)(5\varepsilon+\sqrt{1+24\varepsilon^2})}{(\varepsilon+1)^2}\kappa. \quad (5.14b)$$

Because, as follows from Eq. (5.3), the moduli of $\alpha^{(1)}$ and $\alpha^{(2)}$ should be unity we find from Eqs. (5.14) that the value of $\varepsilon$ has to be zero,

$$\varepsilon=0. \quad (5.15)$$

In this case,

$$\alpha^{(1)} = \kappa, \quad \alpha^{(2)} = -\kappa. \tag{5.16}$$

It is easy to verify from Table III that these values indeed lead to CPT from state $\psi_{-2}$ to state $\psi_2$.

For $\varepsilon = 0$ the MS Rabi frequencies become

$$\Omega^{(1)} = \sqrt{\frac{2}{5}}\Omega, \quad \Omega^{(2)} = \sqrt{\frac{3}{10}}\Omega \quad (2 \leftrightarrow 1), \tag{5.17a}$$

$$\Omega^{(1)} = \sqrt{\frac{2}{3}}\Omega, \quad \Omega^{(2)} = \sqrt{\frac{1}{6}}\Omega \quad (2 \leftrightarrow 2). \tag{5.17b}$$

It follows from Eq. (5.6a) that the same relations hold for the pulse areas $\mathcal{A}^{(1)}$ and $\mathcal{A}^{(2)}$, with $\Omega$ replaced by $\mathcal{A}$.

We note that in the $(2 \leftrightarrow 1)$ case the MS Rabi frequencies are incommensurable, whereas in the $(2 \leftrightarrow 2)$ case they are commensurable, $\Omega^{(1)} = 2\Omega^{(2)}$.

### *2. Exact resonance*

On exact resonance, the phase factor (3.10) is unity, $\kappa = 1$. Therefore, we conclude that conditions (5.16) require us to find pairs $(m^{(1)}, m^{(2)})$ for which $\mathcal{A}^{(1)}$ is an even multiple of $2\pi$ and $\mathcal{A}^{(2)}$ is an odd multiple of $2\pi$, i.e., even $m^{(1)}$ and odd $m^{(2)}$ $(m^{(1)} > m^{(2)})$.

First, we consider the case $J_a = 2 \leftrightarrow J_b = 1$. It follows from Eqs. (5.6a) and (5.17a) that

$$2m^{(2)} = \sqrt{3}m^{(1)}. \tag{5.18}$$

Since $\sqrt{3}$ is an irrational number, this equation cannot be solved exactly for integer $m$. However,we can find numerically pairs of even $m^{(1)}$ and odd $m^{(2)}$ that satisfy Eq. (5.18) arbitrarily accurately. Therefore, we can find approximate CPT in principle, but the required pulse area may be quite large. Such an example can be seen in the upper frame of Fig. 4, where $P_2$ approaches unity for $\mathcal{A} \approx 80$.

We now consider the second case of a $M$ system, $J_a = 2 \leftrightarrow J_b = 2$. Because $\Omega^{(1)} = 2\Omega^{(2)}$ (and hence $\mathcal{A}^{(1)} = 2\mathcal{A}^{(2)}$) it follows from Eq. (5.6b) that

$$m^{(1)} = 2m^{(2)}. \tag{5.19}$$

Therefore condition (5.16) can be satisfied by taking an arbitrary odd $m^{(2)}$ and even $m^{(1)} = 2m^{(2)}$, and then calculate $\mathcal{A}$ and $\varepsilon$ from Eqs (5.8) for these $m^{(1)}$ and $m^{(2)}$: $\varepsilon = 0$, $\mathcal{A} = 2\pi m^{(2)}\sqrt{6}$. This choice of parameters will lead to CPT from state $\psi_{-2}$ to state $\psi_2$. This is an example of a generalized $\pi$ pulse.

Figure 3 displays the population of state $\psi_{+2}$ as a function of the ellipticity $\varepsilon$ and the pulse area $\mathcal{A}$, for $J_a = 2 \leftrightarrow J_b = 1$ (upper frame) and $J_a = 2 \leftrightarrow J_b = 2$ (lower frame). Figure 4 shows the corresponding frames for linear polarization ($\varepsilon = 0$). Three cases of exact CPT are observed in the latter case ($J_a = 2 \leftrightarrow J_b = 2$), whereas only approximate CPT is seen in the former case ($J_a = 2 \leftrightarrow J_b = 1$), in complete agreement with the analytic conclusions above.

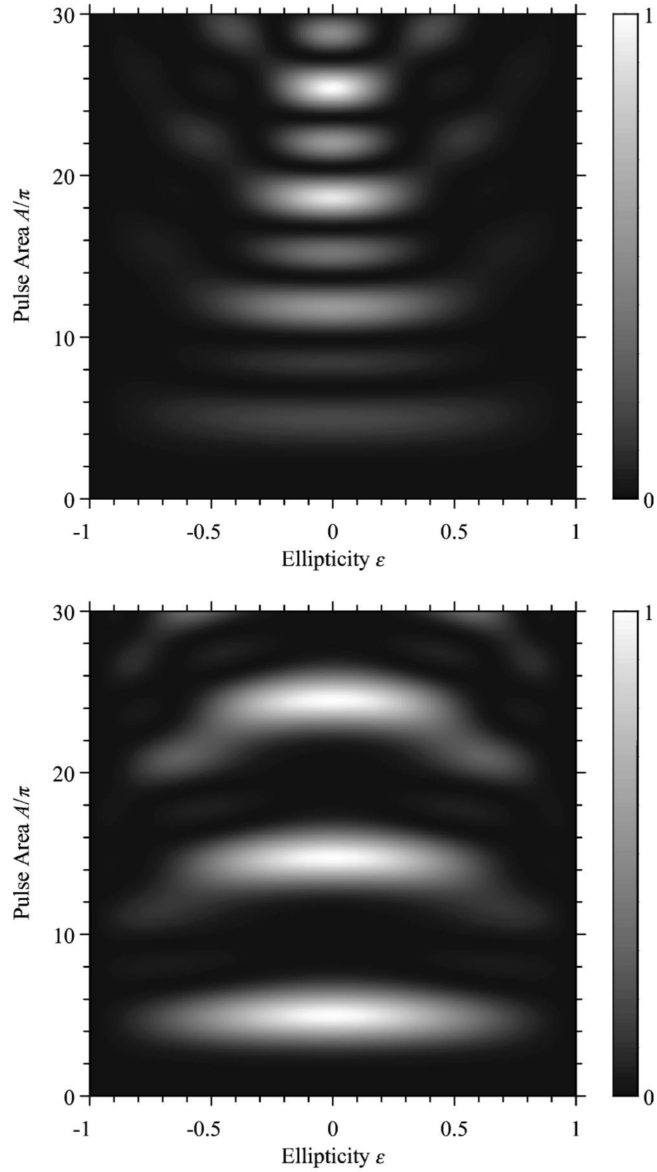


FIG. 3. Population of state $\psi_{+2}$ vs the ellipticity $\varepsilon$ and the pulse area $\mathcal{A}$ in the $M$ system for resonant pulse and linear polarization, $\varepsilon = 0$. The upper frame is for $J_a = 2 \leftrightarrow J_b = 1$ and the lower for $J_a = 2 \leftrightarrow J_b = 2$. Complete population transfer from state $\psi_{-2}$ to state $\psi_2$ occurs only in the latter case, for $\mathcal{A} = 2\pi m^{(2)}\sqrt{6}$ for $m^{(2)} = 1,3,5$. Only approximate CPT can occur in the $J_a = 2 \leftrightarrow J_b = 1$ case due to the incommensurability of the oscillations in the MS basis.

### *3. Off-resonant rectangular pulse*

The reason that exact CPT is impossible for $J_a = 2 \leftrightarrow J_b = 1$ derives from the fact that the Rabi frequencies of the two MS two-state systems (5.17a) are incommensurable. Interestingly, CPT becomes possible off resonance ($\Delta \neq 0$), because then the detuning $\Delta$ provides an additional free parameter.

We illustrate this possibility with the Rabi model, of rectangular pulse and constant detuning. The phase factor (3.10) for this model is $\kappa = \exp(i\Delta T/2)$. Because $\alpha^{(1)}$ and $\alpha^{(2)}$ in Eqs. (5.16) are real, $\kappa$ must be real too, which leads to the condition

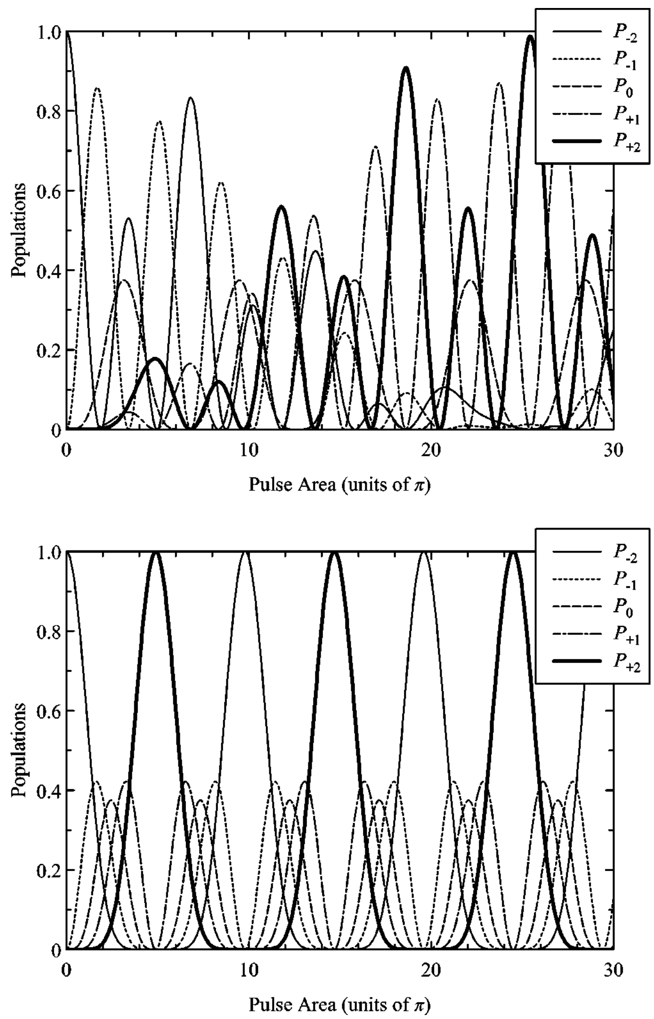


FIG. 4. Populations vs the pulse area $\mathcal{A}$ in the $M$ system for resonant pulse and linear polarization, $\varepsilon=0$. The upper frame is for $J_a=2\leftrightarrow J_b=1$ and the lower for $J_a=2\leftrightarrow J_b=2$. Complete population transfer from state $\psi_{-2}$ to state $\psi_2$ occurs only in the latter case for $\mathcal{A}=2\pi m^{(2)}\sqrt{6}$ for $m^{(2)}=1,3,5$. The irregular (regular) oscillation pattern in the upper (lower) frame is indicative of incommensurate (commensurate) frequencies.

$$\Delta T=2m\pi, \tag{5.20}$$

where $m$ is an integer. Then $\kappa=(-1)^m$ and conditions (5.16) become

$$\alpha^{(1)}=(-1)^m, \quad \alpha^{(2)}=(-1)^{m+1}. \tag{5.21}$$

Because conditions (5.10), when inserted into Eqs. (A7), lead to $\alpha^{(1)}=(-1)^{m^{(1)}}$ and $\alpha^{(2)}=(-1)^{m^{(2)}}$, we should select either odd $m^{(1)}$ and $m$, and even $m^{(2)}$ (case $A$) or even $m^{(1)}$ and $m$, and odd $m^{(2)}$ (case $B$).

By comparing Eq. (5.20) with Eqs. (5.11b) and (5.12b) and taking into account Eq. (5.15) we conclude that the following relations must hold

$$4m^{(2)2}=3m^{(1)2}+m^2 \quad (2\leftrightarrow 1), \tag{5.22a}$$

$$4m^{(2)2}=m^{(1)2}+3m^2 \quad (2\leftrightarrow 2). \tag{5.22b}$$

TABLE IV. Examples of parameter values for which CPT from state $\psi_{-2}$ to $\psi_2$ occurs for off-resonant rectangular pulse (the other parameters are $\varepsilon=0$, $\Delta=2m\pi$). Also solutions are all odd multiples of each set of values.

| | $J_a=2\leftrightarrow J_b=1$ | | | | $J_a=2\leftrightarrow J_b=2$ | | |
|---|---|---|---|---|---|---|---|
| $m^{(1)}$ | $m^{(2)}$ | $m$ | $\Omega T/\pi$ | $m^{(1)}$ | $m^{(2)}$ | $m$ | $\Omega T/\pi$ |
| 8 | 7 | 2 | 24.495 | 22 | 13 | 8 | 50.200 |
| 40 | 37 | 26 | 96.125 | 26 | 19 | 16 | 50.200 |
| 48 | 43 | 22 | 134.907 | 46 | 31 | 24 | 96.125 |
| 80 | 73 | 46 | 206.978 | 74 | 61 | 56 | 118.491 |
| 96 | 91 | 74 | 193.391 | 94 | 49 | 16 | 226.892 |

It follows from these equations that case $A$ is impossible and we can only have even $m^{(1)}$ and $m$, and odd $m^{(2)}$. The CPT value of $\Omega$ is given by

$$\Omega T=2\pi\sqrt{10(m^{(1)2}-m^{(2)2})} \quad (2\leftrightarrow 1), \tag{5.23a}$$

$$\Omega T=2\pi\sqrt{2(m^{(1)2}-m^{(2)2})} \quad (2\leftrightarrow 2), \tag{5.23b}$$

and $\Delta$ is given by Eq. (5.20).

Several sets of parameter values for which CPT from $\psi_{-2}$ to $\psi_2$ occurs are listed in Table IV.

Figure 5 displays the population of state $\psi_{+2}$ as a function of the detuning $\Delta$ and the pulse area $\mathcal{A}$, for $J_a=2\leftrightarrow J_b=1$ (upper frame) and $J_a=2\leftrightarrow J_b=2$ (lower frame). Figure 6 shows the corresponding frames for suitably chosen detunings. One case of CPT is observed in each case, corresponding to the parameters in the first row of Table IV.

The important conclusion is that adding a suitably chosen detuning, Eq. (5.20), makes CPT from state $\psi_{-2}$ to $\psi_2$ possible, while it is impossible on resonance for the case $J_a=2\leftrightarrow J_b=1$ because of the incommensurability of the MS Rabi frequencies $\Omega^{(1)}$ and $\Omega^{(2)}$. Indeed, adding such a detuning allows one to satisfy Eqs. (5.10) and makes the Rabi oscillations commensurate because $\Delta$ changes the frequency of the Rabi oscillations. This fact, that one needs to detune the transition to make CPT possible, is quite intriguing because in a nondegenerate two-state system, CPT is only possible on resonance but not for nonzero detuning.

We emphasize that as far as population transfer from state $\psi_{-2}$ to $\psi_2$ is concerned, the process of stimulated Raman adiabatic passage (STIRAP) [11] provides a superior tool for high efficiency and robustness. Because STIRAP uses delayed pulses, it cannot be treated by the current approach, which requires the same time dependence of all laser pulses.

### D. Complete population transfer between the ground sublevels $\psi_{-2}\rightarrow\psi_0$

It can be shown that CPT from state $\psi_{-2}$ to the middle state of the $M$ system $\psi_0$ is impossible. For $\varepsilon=0$ this is easily seen from the values of the populations listed in Table III. It follows from there that the maximum population state $\psi_0$ can achieve is $\frac{3}{8}$, obtainable for $\alpha^{(1)}=-\kappa$ and $|\kappa|=1$, both for $J_a=2\leftrightarrow J_b=1$ and $J_a=2\leftrightarrow J_b=2$.

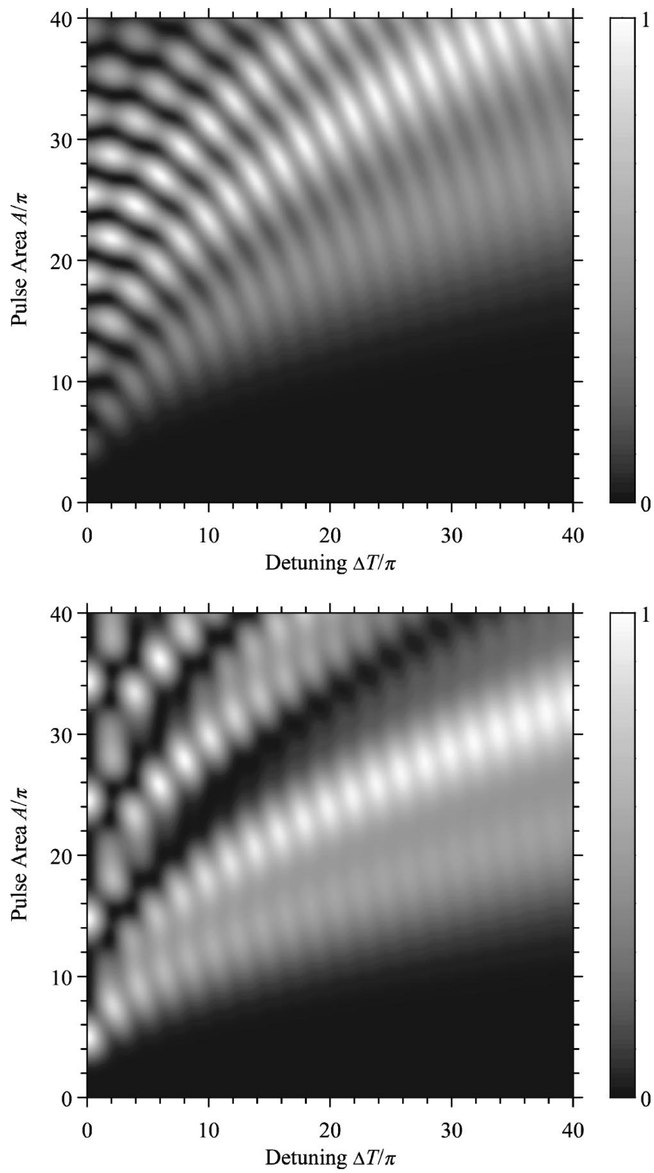


FIG. 5. Population of state $\psi_{+2}$ vs the detuning $\Delta$ and the pulse area $\mathcal{A}$ in the $M$ system for nonresonant rectangular pulse and linear polarization, $\varepsilon=0$. The upper frame is for $J_a=2\leftrightarrow J_b=1$ and the lower for $J_a=2\leftrightarrow J_b=2$. Complete population transfer from state $\psi_{-2}$ to state $\psi_2$ occurs in both cases, for the parameters shown in Table IV.

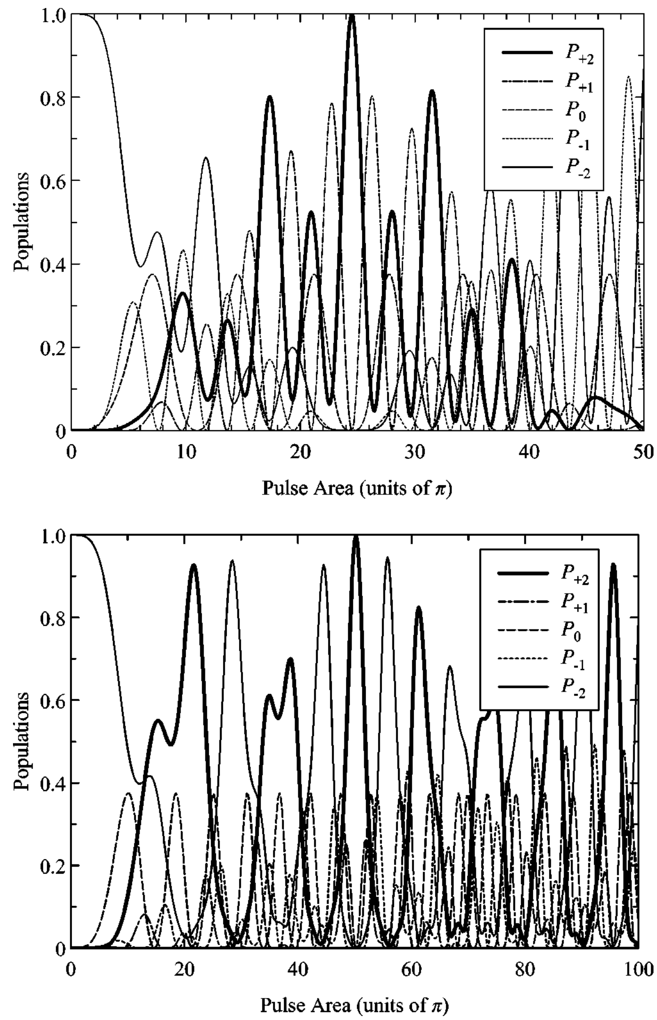


FIG. 6. Populations vs the pulse area $\mathcal{A}$ in the $M$ system for a nonresonant rectangular pulse and linear polarization, $\varepsilon=0$. The upper frame is for $J_a=2\leftrightarrow J_b=1$ and $\Delta T=4\pi$ and the lower for $J_a=2\leftrightarrow J_b=2$ and $\Delta T=16\pi$. Complete population transfer from state $\psi_{-2}$ to state $\psi_2$ occurs in both cases, for the parameters in the first row of Table IV: $\Omega T\approx 24.495\pi$ and $\Omega T\approx 50.200\pi$, respectively.

For nonzero values of the ellipticity, $\varepsilon\neq 0$, the population $P_0$ can reach larger values but never unity. This can be verified by setting $\kappa=\alpha^{(1)}=\alpha^{(2)}=1$ in Eq. (5.1b) and considering $P_0$ as a function of $\varepsilon$. The maximum value of $P_0$ is approximately 0.974 for $J_a=2\leftrightarrow J_b=1$ and 0.676 for $J_a=2\leftrightarrow J_b=2$.

### E. Complete population inversion

A common objective is to induce complete population inversion, that is, to transfer the population from the initial state $\psi_{-2}$ to some combination of the two excited states $\psi_{-1}$ and $\psi_1$. As we have noted at the end of Sec. V A, the population $P_d$ [Eq. (5.2)] associated with the decoupled state $\varphi_d$ is preserved, since the decoupled state does not interact with the laser pulse. Therefore, the population $P_d$ should vanish in order to achieve complete inversion of the system. This condition can be satisfied only for $\varepsilon=1$. Physically this means that the $M$ system cannot be inverted if all states are coupled (i.e., for $|\varepsilon|<1$). If $\varepsilon=1$, only the $\sigma^+$ pulse is present and the $M$ system reduces to a two-state system involving only states $\psi_{-2}$ and $\psi_{-1}$. The population of this system can be inverted using, e.g., an odd-$\pi$ pulse or a chirped adiabatic pulse.

In the other extreme case, $\varepsilon=-1$, only the $\sigma^-$ pulse is present, then $P_d=1$ and the $M$ system, which is initially in state $\psi_{-2}$, does not interact with the laser field.

## VI. THE *W* SYSTEM

Another five-state chainwise-connected system is formed from the magnetic sublevels of two levels with $J_a=1$ or 2 and $J_b=2$ when the system is prepared initially in state $M=-1$ or $M=1$ of the lower level. The treatment of such a system, in a $W$-like linkage pattern, proceeds similarly to the $M$ system, the difference being that now the initial condition is $C_{-1}=1, C_{-2}=C_0=C_1=C_2=0$. There are both similarities and differences from the $M$ system, which we shall discuss below.

### A. Populations in the general case

If the system is initially in state $\psi_{-1}$, the final populations are

$$P_{-1}=|\alpha^{(1)}e_{-}^{(1)2}+\alpha^{(2)}e_{-}^{(2)2}|^2, \quad (6.1a)$$

$$P_{1}=|\alpha^{(1)}e_{-}^{(1)}e_{+}^{(1)}+\alpha^{(2)}e_{-}^{(2)}e_{+}^{(2)}|^2, \quad (6.1b)$$

$$P_{-2}=|\beta^{(1)}c_{-}^{(1)}e_{-}^{(1)}+\beta^{(2)}c_{-}^{(2)}e_{-}^{(2)}|^2, \quad (6.1c)$$

$$P_{0}=|\beta^{(1)}c_{0}^{(1)}e_{-}^{(1)}+\beta^{(2)}c_{0}^{(2)}e_{-}^{(2)}|^2, \quad (6.1d)$$

$$P_{2}=|\beta^{(1)}c_{+}^{(1)}e_{-}^{(1)}+\beta^{(2)}c_{+}^{(2)}e_{-}^{(2)}|^2. \quad (6.1e)$$

For linear polarization ($\varepsilon=0$) we summarize the values of these populations in Table V. Note that the decoupled state $\varphi_d$ does not play any role here, since in this case it is a superposition of the upper levels; therefore, it cannot be populated in the course of the excitation process.

Equations (6.1), along with the analytic formulas for the two-state evolution-matrix elements $\alpha^{(n)}$ and $\beta^{(n)}$, given in the Appendix, provide a number of analytic solutions for the $W$ system. We discuss below some of the ensuing general properties of this system.

### B. All population in the ground levels

All population will be confined to the ground sublevels $\psi_{-1}$ and $\psi_1$ if the upper-level populations vanish, $P_{-2}=P_0=P_2=0$. Because states $\psi_{-1}$ and $\psi_1$ participate only in the MS states $\varphi_e^{(1)}$ and $\varphi_e^{(2)}$, the confinement of the population to states $\psi_{-1}$ and $\psi_1$ is equivalent to requiring that the transition probabilities in the MS basis are zero, i.e.,

$$\beta^{(1)}=\beta^{(2)}=0. \quad (6.2)$$

We can proceed in the same way as for the $M$ system in Sec. V. On resonance, for example, the conditions for no transition to the upper states are given by Eqs. (5.6), the solutions for which in terms of the pulse area $A$ and the ellipticity $\varepsilon$ are given by Eqs. (5.7) (for $J_a=1\leftrightarrow J_b=2$) and by Eqs. (5.8) (for $J_a=2\leftrightarrow J_b=2$). The even integers $k^{(1)}$ and $k^{(2)}$ are arbitrary as long as they satisfy the restrictions (5.9).

The probability amplitudes in the final coherent superposition state are given by

$$C_{-1}(\infty)=\frac{1}{2}(\alpha^{(1)}+\alpha^{(2)})+\frac{5}{2}\frac{\varepsilon(\alpha^{(1)}-\alpha^{(2)})}{\sqrt{1+24\varepsilon^2}}, \quad (6.3a)$$

TABLE V. Final populations for the $W$ system for initial state $\psi_{-1}$ and linear polarization ($\varepsilon=0$).

| | $J_a=2\leftrightarrow J_b=1$ | $J_a=2\leftrightarrow J_b=2$ |
|---|---|---|
| $P_{-1}$ | $\frac{1}{4}\vert\alpha^{(1)}+\alpha^{(2)}\vert^2$ | $\frac{1}{4}\vert\alpha^{(1)}+\alpha^{(2)}\vert^2$ |
| $P_1$ | $\frac{1}{4}\vert\alpha^{(1)}-\alpha^{(2)}\vert^2$ | $\frac{1}{4}\vert\alpha^{(1)}-\alpha^{(2)}\vert^2$ |
| $P_{-2}$ | $\left\vert\frac{\sqrt{5}}{4}\beta^{(1)}-\frac{1}{2}\beta^{(2)}\right\vert^2$ | $\vert\frac{1}{4}\beta^{(1)}+\frac{1}{2}\beta^{(2)}\vert^2$ |
| $P_0$ | $\frac{1}{8}\vert\beta^{(1)}\vert^2$ | $\frac{3}{8}\vert\beta^{(1)}\vert^2$ |
| $P_2$ | $\left\vert\frac{\sqrt{5}}{4}\beta^{(1)}+\frac{1}{2}\beta^{(2)}\right\vert^2$ | $\vert\frac{1}{4}\beta^{(1)}+\frac{1}{2}\beta^{(2)}\vert^2$ |

$$C_{1}(\infty)=\frac{1}{2}\frac{\sqrt{1-\varepsilon^2}}{\sqrt{1+24\varepsilon^2}}(\alpha^{(1)}-\alpha^{(2)})e^{-i\phi}. \quad (6.3b)$$

### C. Complete population transfer between states $\psi_{-1}$ and $\psi_1$

To achieve complete population transfer from state $\psi_{-1}$ to $\psi_1$ it is necessary, in addition to conditions (6.2), that the probability amplitude $C_{-1}(\infty)$ in Eq. (6.3a) should also vanish. By taking into account that the moduli of $\alpha^{(1)}$ and $\alpha^{(2)}$ are unity we find that the only way to satisfy the condition $C_{-1}=0$ is to set $\varepsilon=0$. Then it follows easily from Table V and Eq. (6.3a) that

$$\alpha^{(1)}=-\alpha^{(2)}. \quad (6.4)$$

For example, we can have either of

$$\alpha^{(1)}=1, \quad \alpha^{(2)}=-1, \quad (6.5a)$$

$$\alpha^{(1)}=-1, \quad \alpha^{(2)}=1. \quad (6.5b)$$

Conditions (6.5a) are the same as for the $M$ system in the case of resonance ($\Delta=0$, hence $\kappa=1$), Eqs. (5.16). Therefore, all ensuing conclusions in Secs. V C 2 and V C 3 remain valid. In particular, CPT of $\psi_{-1}\rightarrow\psi_1$ is impossible for resonant excitation in the $J_a=1\leftrightarrow J_b=2$ case, while it is possible for $J_a=2\leftrightarrow J_b=2$; the reason is found again in the commensurability of the MS Rabi frequencies. Off resonance, CPT becomes possible also for $J_a=1\leftrightarrow J_b=2$, for the reasons discussed in Sec. V C 3. The presence of the alternative condition (6.5b), which is opposite to condition (6.5a), provides more possibilities to achieve CPT $\psi_{-1}\rightarrow\psi_1$. However, the conclusions remain qualitatively the same: exact CPT for $J_a=1\leftrightarrow J_b=2$ is impossible for a resonant pulse, whereas it is possible for an off-resonance one.

### D. Complete population inversion

We now address the problem of population inversion, that is, the transfer of all population to the sublevels $\psi_{-2}$, $\psi_0$, and $\psi_2$ of the degenerate upper level. The populations of states $\psi_{-1}$ and $\psi_1$ vanish when

$$\alpha^{(1)}=\alpha^{(2)}=0, \quad (6.6)$$

that is both MS two-state systems should be inverted. Thus unlike the $M$ system, the $W$ system can be inverted even when all states are coupled (i.e., for $\varepsilon \neq \pm 1$) because then the decoupled state is a superposition of excited states and hence it remains unpopulated.

#### 1. Generalized $\pi$ pulses

Condition (6.6) means that the transition probabilities in both MS two-state systems should be equal to unity. This can be achieved easily in the on-resonance case if the pulse areas $\mathcal{A}^{(1)}$ and $\mathcal{A}^{(2)}$, Eqs. (5.6), are both odd multiples of $\pi$, that is,

$$k^{(n)} = 2m^{(n)} + 1, \quad (n = 1,2), \tag{6.7}$$

where $m^{(n)}$ are arbitrary integers.

These conditions can be satisfied for an infinite number of pairs $(\mathcal{A}, \varepsilon)$ of pulse areas and ellipticities, which can be obtained from Eqs. (5.7) and (5.8) with odd $k^{(n)}$ that obey the restrictions (5.9). Table VI lists a few examples of parameter values for which population inversion $\psi_{-1} \rightarrow [\psi_{-2}, \psi_0, \psi_2]$ occurs. Figure 7 illustrates two of these examples for the parameters in the first row of Table VI.

#### 2. Chirped pulses

Apart from resonant odd-$\pi$ pulses, conditions (6.6), which require CPT in each of the two MS two-state systems, cannot be satisfied for pulses with constant detuning, such as the rectangular pulse in the Rabi model and the hyperbolic-secant pulse in the Rosen-Zener model. These conditions can be satisfied, however, for chirped pulses, such as the hyperbolic-secant pulse with the hyperbolic-tangent chirp in the Allen-Eberly-Hioe model and the constant pulse with linear detuning in the Landau-Zener model.

In excitation with chirped pulses it is not the completeness (100%) of the population transfer that is significant but rather the robustness of the (high) transfer efficiency against variations in the interaction parameters, which derives from the adiabatic nature of the population transfer. Typically, if the Rabi frequency $\Omega$ and the chirp rate $C$ are sufficiently large compared to the inverse pulse width $1/T$, the transfer efficiency remains little changed for small-to-moderate variations in $\Omega$ and $C$. Because the MS Rabi frequencies in the two MS two-state systems are different and because the chirp rate in both is the same (since the detuning $\Delta(t)$ is the same), it is the smaller of the two MS Rabi frequencies, $\Omega^{(2)}$, that determines when the transfer efficiency will exceed the value

$$\mathcal{P} \gtrsim 1 - \epsilon, \tag{6.8}$$

where $\epsilon$ is the deviation from CPT. We consider for simplicity only the case when the ellipticity is zero, $\varepsilon = 0$; then the MS Rabi frequencies are given by Eqs. (5.17).

For the Landau-Zener model [Eqs. (A9)], Eq. (6.8) leads to

$$\Omega^2 \gtrsim \frac{20|C|}{3\pi} |\ln \epsilon| \quad (1 \leftrightarrow 2), \tag{6.9a}$$

$$\Omega^2 \gtrsim \frac{12|C|}{\pi} |\ln \epsilon| \quad (2 \leftrightarrow 2). \tag{6.9b}$$

TABLE VI. Examples of parameter values for which complete population transfer occurs from sublevel $\psi_{-1}$ of the ground level to the sublevels $\psi_{-2}$, $\psi_0$, and $\psi_2$ of the excited level.

| | $J_a=2 \leftrightarrow J_b=1$ | | | | $J_a=2 \leftrightarrow J_b=2$ | | |
|---|---|---|---|---|---|---|---|
| $k^{(1)}$ | $k^{(2)}$ | $\mathcal{A}/\pi$ | $\varepsilon$ | $k^{(1)}$ | $k^{(2)}$ | $\mathcal{A}/\pi$ | $\varepsilon$ |
| 5 | 3 | 6.969 | $\pm 0.641$ | 5 | 3 | 6.387 | $\pm 0.658$ |
| 7 | 3 | 9.103 | $\pm 0.964$ | 7 | 5 | 9.423 | $\pm 0.892$ |
| 7 | 5 | 10.282 | $\pm 0.416$ | 9 | 5 | 11.278 | $\pm 0.503$ |
| 9 | 5 | 12.306 | $\pm 0.727$ | 9 | 7 | 12.490 | $\pm 0.967$ |
| 9 | 7 | 13.628 | $\pm 0.286$ | 11 | 7 | 14.283 | $\pm 0.751$ |

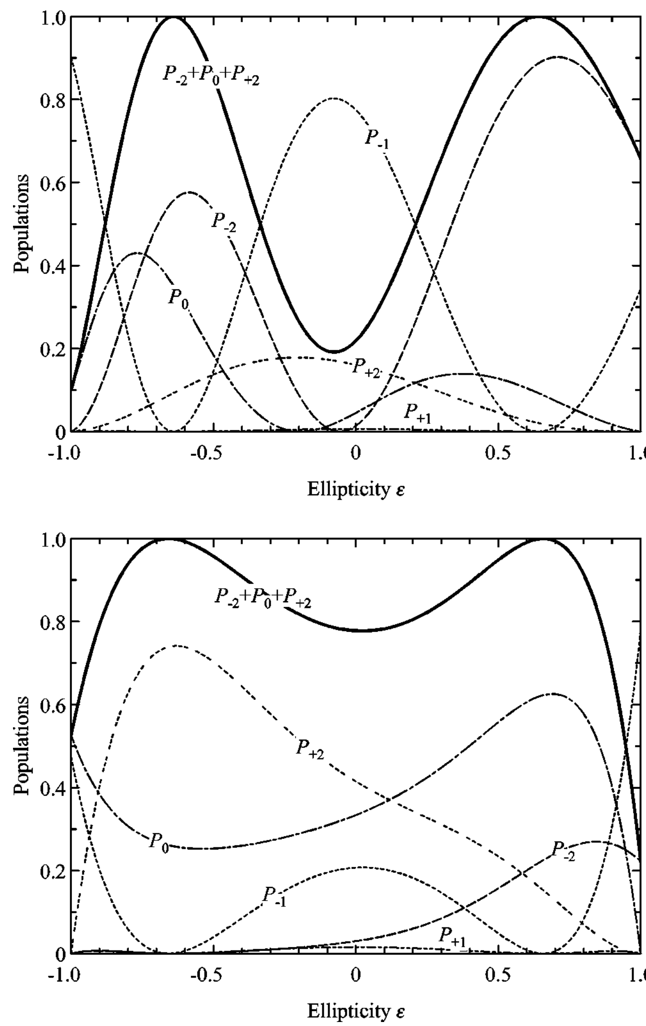


FIG. 7. Populations vs the ellipticity $\varepsilon$ in the $W$ system for resonant excitation $(\Delta = 0)$. The upper frame is for $J_a = 1 \leftrightarrow J_b = 2$ and pulse area $\mathcal{A} = \sqrt{340/7}\pi \approx 6.969\pi$ and the lower for $J_a = 2 \leftrightarrow J_b = 2$ and pulse area $\mathcal{A} = \sqrt{204/5}\pi \approx 6.387\pi$. Complete population transfer from state $\psi_{-1}$ to the upper sublevels $\psi_{-2}$, $\psi_0$ and $\psi_2$ occurs in both cases, for the parameters in the first row of Table VI: $\varepsilon \approx \pm 0.641$ and $\varepsilon \approx \pm 0.658$, respectively.

For the Allen-Eberly-Hioe model [Eqs. (A16)], Eq. (6.8) reduces to

$$\sqrt{\frac{3}{10}}\Omega \gtrsim |B| \gtrsim \frac{1}{\pi T}\ln\left(\frac{1+\sqrt{1-\epsilon}}{\epsilon}\right) \quad (1\leftrightarrow 2), \tag{6.10a}$$

$$\sqrt{\frac{1}{6}}\Omega \gtrsim |B| \gtrsim \frac{1}{\pi T}\ln\left(\frac{1+\sqrt{1-\epsilon}}{\epsilon}\right) \quad (2\leftrightarrow 2). \tag{6.10b}$$

## VII. SUMMARY AND CONCLUSIONS

We have discussed the extension of the MS transformation to time-dependent pulses, restricted only by the condition that, in addition to having only two distinct (but possibly time-dependent) detunings, all of the pulse envelopes (Rabi frequencies) have the same time dependence. The MS transformation converts the full linkage pattern into separate two-state systems. In a number of cases (i.e., particular choices for the time dependences of the Rabi frequency and the detuning) there exist analytic expressions for the two independent elements of the transition matrix. We have used these, together with the time-independent MS transformation, to evaluate the conditions needed to produce such desired results as complete population transfer. We have illustrated the application of this technique to several two-state models, including chirped pulses.

We have used this analytic technique to study the properties of the population dynamics in chainwise-connected five-state systems in *M*- and *W*-linkage patterns. Such systems are formed amongst the magnetic sublevels in degenerate two-level systems with angular momenta $J=1$ or 2, driven by elliptically polarized laser pulse. We have derived various conditions for complete population transfer between the magnetic sublevels, with examples of generalized $\pi$ pulses and chirped pulses.

## ACKNOWLEDGMENTS

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## APPENDIX: ANALYTICALLY SOLUBLE TWO-STATE MODELS

We collect here expressions for the elements of the transition matrix

$$\bar{\mathsf{S}} = \begin{bmatrix} \alpha & -\beta^* \\ \beta & \alpha^* \end{bmatrix}, \tag{A1}$$

for a number of analytically soluble models, defined by the Hamiltonian

$$\bar{\mathsf{H}} = \frac{\hbar}{2}\begin{bmatrix} -\Delta(t) & \Omega(t) \\ \Omega(t) & \Delta(t) \end{bmatrix}. \tag{A2}$$

### 1. Exact resonance

When excitation is resonant, $\Delta_a=\Delta_b=0$, the elements of the two-state transition matrix are expressible, for any pulse shape $\Omega(t)$, in terms of the pulse area

$$\mathcal{A} = \int_{-\infty}^{+\infty}\Omega(t)dt, \tag{A3}$$

as simply

$$\alpha = \cos(\mathcal{A}/2), \tag{A4a}$$

$$\beta = -i\sin(\mathcal{A}/2). \tag{A4b}$$

The transition probability is

$$\mathcal{P} = |\beta|^2 = \sin^2(\mathcal{A}/2). \tag{A5}$$

Complete population transfer $(\alpha=0,|\beta|=1)$ occurs when $\mathcal{A}=k\pi$ for any odd integer $k$ (odd-$\pi$ pulse). Complete population return $(|\alpha|=1,\beta=0)$, occurs when $\mathcal{A}=k\pi$ for any even integer $k$ (even-$\pi$ pulse).

Models with nonzero detuning typically introduce a change of independent variable from time $t$ to some appropriately chosen function $z(t)$. Such a transformation can be used to transform the two-state Schrödinger equation into a second-order equation satisfied by one of the many special functions of mathematical physics, cf. Ref. [4]. We shall summarize below the best known nonresonant analytic solutions.

### 2. The Rabi model

The simplest and most widely used soluble model of a two-state system with nonresonant excitation is the Rabi model, with rectangular pulse shape and constant detuning,

$$\Omega(t) = \Omega_0 \quad (-T/2 \leqslant t \leqslant T/2), \tag{A6a}$$

$$\Delta(t) = \Delta_0. \tag{A6b}$$

In this model the elements of the transfer matrix (3.9) are given by

$$\alpha = \cos(\tfrac{1}{2}\tilde{\Omega}T) + i\frac{\Delta}{\tilde{\Omega}}\sin(\tfrac{1}{2}\tilde{\Omega}T), \tag{A7a}$$

$$\beta = -i\frac{\Omega}{\tilde{\Omega}}\sin(\tfrac{1}{2}\tilde{\Omega}T), \tag{A7b}$$

where $\tilde{\Omega}=\sqrt{\Omega_0^2+\Delta_0^2}$. The transition probability is

$$\mathcal{P}=|\beta|^2=\frac{\Omega_0^2}{\Omega_0^2+\Delta_0^2}\sin^2\left(\tfrac{1}{2}\sqrt{\Omega_0^2+\Delta_0^2}T\right), \tag{A8}$$

The transition probability is always less than unity ($|\beta|<1,|\alpha|>0$), unless $\Delta_0=0$, and has infinitely many zeroes ($|\alpha|=1,\beta=0$).

### 3. The Landau-Zener model

In the Landau-Zener model [17], the Rabi frequency is constant and the detuning varies linearly with time (linear frequency chirp),

$$\Omega(t)=\Omega_0, \tag{A9a}$$

$$\Delta(t)=Ct. \tag{A9b}$$

The coupling $\Omega(t)$ is supposed to last from $t\to-\infty$ to $t\to+\infty$. The transition probability is

$$\mathcal{P}=|\beta|^2=1-e^{-\pi\Lambda}, \tag{A10}$$

where

$$\Lambda=\frac{\Omega_0^2}{2C}. \tag{A11}$$

The transition probability is always nonzero (which means that $|\alpha|<1,|\beta|>0$), unless $\Omega_0=0$, and approaches unity as $\Omega_0$ increases.

### 4. The Demkov-Kunike Model

#### *a. General case*

The Demkov-Kunike (DK) model [18] is defined by

$$\Omega(t)=\Omega_0\,\mathrm{sech}(t/T), \tag{A12a}$$

$$\Delta(t)=\Delta_0+B\tanh(t/T). \tag{A12b}$$

The pulse area is $\mathcal{A}=\pi\Omega_0 T$. The transition probability is

$$\mathcal{P}=|\beta|^2=\frac{\cosh(\pi BT)-\cos\left(\pi T\sqrt{\Omega_0^2-B^2}\right)}{\cosh(\pi\Delta_0 T)+\cosh(\pi BT)}. \tag{A13}$$

The DK model has several important particular cases.

#### *b. The Rosen-Zener Model*

The Rosen-Zener model [19] is a particular case of the DK model for constant detuning ($B=0$),

$$\Omega(t)=\Omega_0\,\mathrm{sech}(t/T), \tag{A14a}$$

$$\Delta(t)=\Delta_0. \tag{A14b}$$

The transition probability is

$$\mathcal{P}=|\beta|^2=\frac{\sin^2\left(\tfrac{1}{2}\pi\Omega_0 T\right)}{\cosh^2\left(\tfrac{1}{2}\pi\Delta_0 T\right)}. \tag{A15}$$

It is always less than unity (which means that $|\alpha|>0,|\beta|<1$), unless $\Delta_0=0$, and vanishes for even-$\pi$ pulses ($\mathcal{A}=2m\pi$), as for the exact-resonance solution.

#### *c. The Allen-Eberly-Hioe model*

The Allen-Eberly-Hioe model [1,20] is a particular case of the DK model for $\Delta_0=0$,

$$\Omega(t)=\Omega_0\,\mathrm{sech}(t/T), \tag{A16a}$$

$$\Delta(t)=B\tanh(t/T). \tag{A16b}$$

The transition probability is

$$\mathcal{P}=|\beta|^2=1-\frac{\cos^2\left(\tfrac{1}{2}\pi T\sqrt{\Omega_0^2-B^2}\right)}{\cosh^2\left(\tfrac{1}{2}\pi BT\right)}. \tag{A17}$$

It is always nonzero (which means that $|\alpha|<1,|\beta|>0$), unless $\Omega_0=0$, and approaches rapidly unity as $B$ increases (provided $\Omega_0>B$).

#### *d. The Bambini-Berman model*

The Bambini-Berman model [21] is defined by

$$\Omega(t)=\Omega_0\,\mathrm{sech}(t/T), \tag{A18a}$$

$$\Delta(t)=B[1+\tanh(t/T)]. \tag{A18b}$$

The transition probability is

$$\mathcal{P}=|\beta|^2=\frac{1}{2}-\frac{1}{2}\,\frac{\cos\left(\pi T\sqrt{\Omega_0^2-B^2}\right)}{\cosh(\pi BT)}. \tag{A19}$$

It never reaches zero or unity (which means that $|\alpha|\neq 0,1;|\beta|\neq 0,1$), unless $\Omega_0=0$ or $B=0$, and approaches $\frac{1}{2}$ as $B$ increases (provided $\Omega_0>B$).


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