Complete population transfer in a degenerate three-state atom

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We have found conditions required to achieve complete population transfer, via coherent population trapping, from an initial state to a designated final state at a designated time in a degenerate three-state atom, where transitions are caused by an external interaction. Complete population transfer from an initially occupied state one to a designated state two occurs under two conditions. First, there is a constraint on the ratios of the transition matrix elements of the external interaction. Second, there is a constraint on the action integral over the interaction, or "area," corresponding to the phase shift induced by the external interaction. Both conditions may be expressed in terms of simple odd integers. Some specific examples are discussed.

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I. INTRODUCTION

Population control in quantum systems, namely, transfer of electrons from an ensemble of atoms all in the same initial state to specified final states, is used in problems ranging from coherent population trapping [1–5], including electromagnetically induced transparency [6–10], and quantum computing [11–17], to chemical dynamics [18–21]. These problems are modeled in terms of an *n*-state (often three-state) atom interacting with a strong external field [22–27]. In this paper we consider population control in a nearly degenerate three-state atom. We show that the three-state atom is relatively easy to understand in the degenerate limit, where all three states have the same energy. We present the conditions required to achieve complete population transfer in a degenerate three-state atom where the matrix elements of the external interaction have a common time dependence.

Our degenerate approximation [28] is mathematically similar to the rotating wave approximation (RWA) [22,25], which has been widely applied to both two- and three-state atomic models. However, in the RWA degenerate atomic states are not used. Instead one tunes the frequency of the external field to the frequency difference of two nondegenerate states so that the detuning parameter [23] tends to zero. Thus, in RWA the initial state of an atom plus one photon is degenerate in energy with the final state of the atom. One advantage of using degenerate atomic states, as done in this paper, is that one may use external interactions with a broad range of frequencies. Another advantage is that the interaction frequency can be used as a control parameter, e.g., to vary the duration of time that the transferred population remains in the designated state, or to reduce the population leakage that occurs when the three states are not fully degenerate.

In the following section we derive analytic formulas for the probabilities, $P_{1,2,3}(t)$, that the electron is in state one, two, or three at time *t*. Then we seek conditions for complete population transfer to a designated state that is initially unoccupied at a designated time t_0. This places conditions on both the relative strengths of the interaction matrix elements, $V_{ij}(t)$, and the action integral, $A(t_0)=\int_0^{t_0}V(t')dt'$. We show that these conditions for complete transfer may be expressed in terms of two odd integers, n_1 and n_2. Some analysis is presented for population leakage that occurs when the states are not quite degenerate. Calculations using dipole allowed transitions are presented.

II. THEORY

Let us consider an *n*-state atom interacting with an external field, $V_{ext}(\vec{r},t)$. The total Hamiltonian for this system is $H=H_0+V_{ext}(\vec{r},t)$. The *n* eigenstates, ϕ_k, and corresponding eigenenergies, E_k, of H_0 are assumed to be known. The total wave function may be expanded in terms of the known eigenstates, namely, $\Psi(t)=a_1(t)\phi_1+a_2(t)\phi_2+\cdots+a_n(t)\phi_n$. With atomic units, using $i\dot{\Psi}=(H_0+V_{ext}(\vec{r},t))\Psi$, with $H_0\phi_k=E_k\phi_k$ and $\int\phi_j^*\phi_k d\vec{r}=\delta_{jk}$, one then obtains [23]

$$i\dot{a}_j(t)=E_j a_j(t)+\sum_{k=1}^{n} V_{jk}(t)a_k(t), \tag{1}$$

where $V_{jk}(t)=\int\phi_j^* V_{ext}(\vec{r},t)\phi_k d\vec{r}$. These equations are exact for an *n*-state atom.

We now require that the system be degenerate, namely that all the energies E_j are the same. Since the zero point of energy is arbitrary, one may generally set $E_j=0$. These conditions give the coupled equations for a degenerate *n*-state system, namely,

$$i\dot{a}_j(t)=\sum_{k=1}^{n} V_{jk}(t)a_k(t). \tag{2}$$

We use the initial conditions $a_1(0)=1$, and $a_j(0)=0$ for $j\neq 1$. We additionally require that all of the $V_{jk}(t)$ have the same time dependence. Here we also take $V_{jk}=V_{kj}$ to be real.

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A. Degenerate three-state atom

The degenerate three-state atom can be solved analytically. Recalling that the $V_{jk}(t)$ have a common time dependence, we choose $V(t)=V_{23}(t)=\alpha^{-1}V_{12}(t)=\beta^{-1}V_{13}(t)=\epsilon_j^{-1}V_{jj}(t)$. A relatively simple solution, presented next, is found by taking $\epsilon_j=0$. More general solutions for $\epsilon_j\neq 0$ are discussed in the Appendix. With $\epsilon_j=0$ and $n=3$, Eq. (2) becomes

$$i\dot{a}_1(t)=\alpha V(t)a_2(t)+\beta V(t)a_3(t),$$

$$i\dot{a}_2(t)=\alpha V(t)a_1(t)+V(t)a_3(t),$$

$$i\dot{a}_3(t)=\beta V(t)a_1(t)+V(t)a_2(t). \tag{3}$$

Now consider the linear combination $c(t)=a_1(t)+xa_2(t)+ya_3(t)$, where x and y are some time-independent coefficients that we next determine. Then,

$$i\dot{c}(t)=i\dot{a}_1(t)+xi\dot{a}_2(t)+yi\dot{a}_3(t)=\alpha V(t)a_2(t)+\beta V(t)a_3(t)+x[\alpha V(t)a_1(t)+V(t)a_3(t)]+y[\beta V(t)a_1(t)+V(t)a_2(t)]. \tag{4}$$

Set $z=\alpha x+\beta y$. Then, $i\dot{c}(t)=zV(t)[a_1(t)+(\alpha+y)/(\alpha x+\beta y)a_2(t)+(\beta+x)/(\alpha x+\beta y)a_3(t)]$. We require that $i\dot{c}(t)=zV(t)c(t)$. This holds if and only if $x=(\alpha+y)/(\alpha x+\beta y)$ and $y=(\beta+x)/(\alpha x+\beta y)$. After some algebra this leads to the useful cubic equation,

$$(\beta^2-\alpha^2)y^3+\alpha(2-\alpha^2-\beta^2)y^2+(2\alpha^2-\beta^2-1)y+\alpha(\beta^2-1)=0. \tag{5}$$

This determines three sets of eigenvalues, $\{x_j\}$ and $\{y_j\}$, and three eigenfunctions, $c_j(t)=e^{-iz_jA(t)}$, where $A(t)=\int_0^t V(t')dt'$. Specifically, $c_j=\Sigma_{i=1}^3\mathcal{M}_{ji}a_i$, where

$$\mathcal{M}=\begin{pmatrix}1 & x_1 & y_1\\ 1 & x_2 & y_2\\ 1 & x_3 & y_3\end{pmatrix}. \tag{6}$$

This matrix, $\mathcal{M}$, may be inverted, namely,

$$\mathcal{M}^{-1}=\frac{1}{\Delta}\begin{pmatrix}x_2y_3-x_3y_2 & x_3y_1-x_1y_3 & x_1y_2-x_2y_1\\ y_2-y_3 & y_3-y_1 & y_1-y_2\\ x_3-x_2 & x_1-x_3 & x_2-x_1\end{pmatrix}, \tag{7}$$

where $\Delta=\det(\mathcal{M})=x_1y_2+x_2y_3+x_3y_1-x_1y_3-x_2y_1-x_3y_2$.

Now one may express the unperturbed state amplitudes, $a_j(t)$, in terms of the dressed-state amplitudes, $c_j(t)$. Using $c_j(t)=e^{-iz_jA(t)}$, one has $a_i(t)=\Sigma_{j=1}^3\mathcal{M}_{ij}^{-1}c_j(t)=\Sigma_{j=1}^3\mathcal{M}_{ij}^{-1}e^{-iz_jA(t)}$. This leads to

$$P_1(t)=|a_1(t)|^2=\frac{1}{\Delta^2}[(x_2y_3-x_3y_2)^2+(x_3y_1-x_1y_3)^2+(x_1y_2-x_2y_1)^2+2(x_2y_3-x_3y_2)(x_3y_1-x_1y_3)\cos((z_1-z_2)A(t))+2(x_2y_3-x_3y_2)(x_1y_2-x_2y_1)\cos((z_1-z_3)A(t))+2(x_3y_1-x_1y_3)(x_1y_2-x_2y_1)\cos((z_2-z_3)A(t))],$$

$$P_2(t)=|a_2(t)|^2=\frac{1}{\Delta^2}[(y_2-y_3)^2+(y_3-y_1)^2+(y_1-y_2)^2+2(y_2-y_3)(y_3-y_1)\cos((z_1-z_2)A(t))+2(y_2-y_3)(y_1-y_2)\cos((z_1-z_3)A(t))+2(y_3-y_1)(y_1-y_2)\cos((z_2-z_3)A(t))],$$

$$P_3(t)=|a_3(t)|^2=\frac{1}{\Delta^2}[(x_3-x_2)^2+(x_1-x_3)^2+(x_2-x_1)^2+2(x_3-x_2)(x_1-x_3)\cos((z_1-z_2)A(t))+2(x_3-x_2)(x_2-x_1)\cos((z_1-z_3)A(t))+2(x_1-x_3)(x_2-x_1)\cos((z_2-z_3)A(t))]. \tag{8}$$

Equation (8) generally gives the transition probabilities to all three final states for arbitrary $V_{jk}(t)$, and corresponds to the more general expression [29], $P_k(t)=\Sigma_i^n\Sigma_j^n\mathcal{M}_{ki}^{-1}\mathcal{M}_{kj}^{-1}\cos[(z_i-z_j)A(t)]$. Since the x_j's, y_j's, and z_j's vary with the $V_{jk}(t)$, one may then seek conditions on the matrix elements V_{jk} such that the electron populations $P_k(t_0)$ take any desired values at any specified time, $t=t_0$.

B. Complete population transfer

Next we seek conditions on the $V_{jk}(t)$ such that at time t_0 the electron population is completely transferred from its initial state $i=1$ to a different final state, $i=2$ or 3. As shown from Eq. (A7) in Appendix A, the condition that $\epsilon_j=0$ yields $\alpha=2/y-y$ and $\beta=\pm 1$. From the Appendix taking $\beta=+1$ in case (i) and using $r=\pm\sqrt{2/(n_1n_2)}$, one has

$$\{x_1,x_2,x_3\}=\{1,1,-1\},$$

$$\{y_1,y_2,y_3\}=\{y_+,y_-,0\}=\frac{1}{2}\{-\alpha+\sqrt{\alpha^2+8},-\alpha-\sqrt{\alpha^2+8},0\}=r\{n_2,-n_1,0\},$$

$$\{z_1,z_2,z_3\}=\{\alpha+y_+,\alpha+y_-,-\alpha\}=\frac{1}{2}\{\alpha+\sqrt{\alpha^2+8},\alpha-\sqrt{\alpha^2+8},-2\alpha\}=\{-y_-,-y_+,-\alpha\}=r\{n_1,-n_2,n_2-n_1\},$$

TABLE I. Some values for the action integral $A(t_0)$ and the relative interaction strength α, allowed for total population transfer. These values are found using Eq. (10) subject to the conditions listed. The three cases for $\{n_e, n_o'\}$, $\{n_o', n_o\}$, and $\{n_o, n_e\}$ are explained in Appendix A and below Eq. (12).

$n_1 \cdot n_2$	n_1	n_2	n_e k	n_o' $-k'$	n_o' k'	n_o k	n_o $-k$	n_e k'	$A(t_0)$	α
5	± 1	± 5	± 2	± 3	± 3	∓ 1	∓ 1	± 2	± 1.656	∓ 2.530
5	± 5	± 1	± 2	∓ 1	∓ 1	± 3	± 3	± 2	± 1.656	± 2.530
9	± 3	± 3	± 2	± 1	± 1	± 1	± 1	± 2	± 2.221	0.000
11	± 1	± 11	± 4	± 7	± 7	∓ 3	∓ 3	± 4	± 2.456	∓ 4.264
11	± 11	± 1	± 4	∓ 3	∓ 3	± 7	± 7	± 4	± 2.456	± 4.264
17	± 1	± 17	± 6	± 11	± 11	∓ 5	∓ 5	± 6	± 3.053	∓ 5.488
17	± 17	± 1	± 6	∓ 5	∓ 5	± 11	± 11	± 6	± 3.053	± 5.488
27	± 3	± 9	± 4	± 5	± 5	∓ 1	∓ 1	± 4	± 3.848	∓ 1.633
27	± 9	± 3	± 4	∓ 1	∓ 1	± 5	± 5	± 4	± 3.848	± 1.633
35	± 1	± 35	± 12	± 23	± 23	∓ 11	∓ 11	± 12	± 4.381	∓ 8.128

$$\Delta = 2(y_- - y_+) = -2\sqrt{\alpha^2+8} = -2r(n_1+n_2). \tag{9}$$

Here n_1 and n_2 are integers whose values are specified below. This leads to certain allowed values of the action integral $A(t_0)$ and the relative interaction strength α, namely,

$$3rA(t_0) = \pm 3\sqrt{\frac{2}{n_1 n_2}}A(t_0) = \pi,$$

$$\alpha = r(n_1 - n_2). \tag{10}$$

The same allowed values of $A(t_0)$, α, and β are found in all cases, shown in the Appendix.

One may use the values of the x_j, y_j, z_j, and Δ from Eq. (9) in Eq. (8) to obtain explicit expressions for the transition probabilities, namely,

$$P_1(t) = \frac{1}{2(n_1+n_2)^2}\{n_1^2+n_2^2+n_1n_2[1+\cos((n_1+n_2)rA(t))]+(n_1+n_2)[n_1\cos((2n_1-n_2)rA(t))+n_2\cos((2n_2-n_1)rA(t))]\},$$

$$P_2(t) = \frac{1}{2(n_1+n_2)^2}\{n_1^2+n_2^2+n_1n_2[1+\cos((n_1+n_2)rA(t))]-(n_1+n_2)[n_1\cos((2n_1-n_2)rA(t))+n_2\cos((2n_2-n_1)rA(t))]\},$$

$$P_3(t) = \frac{2n_1n_2}{(n_1+n_2)^2}\sin^2\left[\left(\frac{n_1+n_2}{2}\right)rA(t)\right], \tag{11}$$

where n_1, n_2 are odd integers. The constraints on the values of n_1 and n_2 are as follows. The condition $P_1=0$ requires that $\frac{1}{3}(2n_1-n_2)$ and $\frac{1}{3}(2n_2-n_1)$ are both odd integers (i.e., n_0 and n_o'), and also requires that $\frac{1}{3}(n_1+n_2)$ is an even integer. The condition $P_2=1$ imposes the same requirements. The condition $P_3=0$ only requires that $\frac{1}{3}(n_1+n_2)$ be an even integer, but $P_3 \geqslant 0$ requires that $n_1n_2 \geqslant 0$.

Finally a slightly neater solution may now be obtained from the requirements above that $2n_1-n_2=3n_o$ and $2n_2-n_1=3n_o'$, where n_o and n_o' are both odd integers (either positive or negative). It is easily shown that $n_1+n_2=3(n_o+n_o')$ and $n_1n_2=2n_o^2+5n_on_o'+2n_o^2$. Then $r=\pm(n_o^2+\frac{5}{2}n_on_o'+n_o'^2)^{-1/2}$, $\alpha=r(n_o-n_o')$ and $3rA(t_0)=\pi$. Now,

$$P_1(t) = \frac{1}{2(n_1+n_2)^2}\{n_1^2+n_2^2+n_1n_2[1+\cos((n_o+n_o')\pi[A(t)/A(t_0)])]+(n_1+n_2)\times[n_1\cos(n_o\pi[A(t)/A(t_0)])+n_2\cos(n_o'\pi[A(t)/A(t_0)])]\},$$

$$P_2(t) = \frac{1}{2(n_1+n_2)^2}\{n_1^2+n_2^2+n_1n_2[1+\cos((n_o+n_o')\pi[A(t)/A(t_0)])]-(n_1+n_2)\times[n_1\cos(n_o\pi[A(t)/A(t_0)])+n_2\cos(n_o'\pi[A(t)/A(t_0)])]\},$$

$$P_3(t) = \frac{2n_1n_2}{(n_1+n_2)^2}\sin^2(\tfrac{1}{2}(n_o+n_o')\pi[A(t)/A(t_0)]). \tag{12}$$

Here n_o and n_o' are arbitrary odd integers, $n_1=2n_o+n_o'$ and $n_2=n_o+2n_o'$. At $t=t_0$, $A(t)/A(t_0)=1$, so that $P_2(t_0)=1$ with $P_1(t_0)=P_3(t_0)=0$. This yields complete population transfer from state one to state two at $t=t_0$. We regard n_o and n_o' as the more fundamental numbers since they obey the simplest rules.

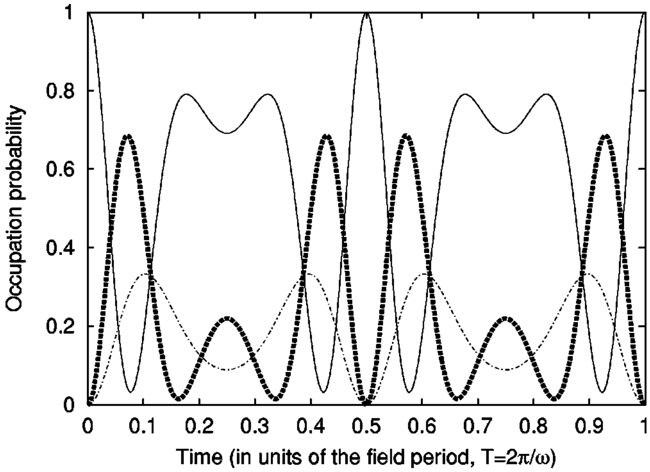

FIG. 1. Occupation probabilities as a function of time over one period, T, of the external field, $V(t)=V_0\cos(\omega t)$. The thick dashed line is the probability $P_2(t)$ that the electron is transferred to the target state, namely, state two. The solid line is the probability $P_1(t)$ that the electron is in state one, the state in which it began initially. The thin dash-dotted line is the probability $P_3(t)$ that the electron is in state three. In this figure $\alpha=V_{12}/V_{23}=2$ and the action area at $t=t_0$ is $A(t_0)=\int_0^{t_0}V(t')dt'=1.5$. Since α and $A(t_0)$ do not correspond to values given by Eq. (10), complete transfer to state two does not occur.

C. Quantum numbers

Some allowed values of the action integral, $A(t_0)$, and the relative interaction strength, α, are given in Table I. This table includes values of $\{n_1,n_2\}$, $\{n_o,n_o'\}$ and all three $\{k,k'\}$ cases defined in the Appendix. From more complete numerical output we confirm that n_1 and n_2 each acquire all possible odd integer values, although values of the product $n_1\cdot n_2$ are restricted. This is consistent with the condition that $n_1=2n_o+n_o'$ and $n_2=n_o+2n_o'$, where n_o and n_o' are arbitrary odd integers. It is also evident that the even integer, $n_e=n_o+n_o'$, takes on all even values. One may also show algebraically that for each value of the even integer k in case (i) there are two odd values of an odd integer k in case (ii) and vice versa. The sets of integers $\{n_1,n_2\}$, $\{n_o,n_o'\}$, and $\{k,k'\}$ are redundant. The three sets of $\{k,k'\}$ correspond to a single set $\{n_1,n_2\}$, while the $\{n_o,n_o'\}$ are in one to one correspondence with the $\{n_1,n_2\}$.

Numerical calculations for the time dependence of the populations in a degenerate three-state atom perturbed by external interactions with $V(t)=V_0\cos(\omega t)$ are presented in Figs. 1–4. These results were obtained by using a standard fourth-order Runge-Kutta numerical integration of Eq. (3) with $\beta=1$ for various values of α and $A(t_0)$. For most values of α the population transfer into either state two or state three is always incomplete. A typical case is shown in Fig. 1 where $\alpha=2$ and $A(t_0)=1.5$. We note that $P_1(t)$ does return to zero twice in each period, T, of the external field, but that complete population transfer to an initially unoccupied state never occurs.

Calculations using a few values of α that permit complete transfer to state two are shown in Figs. 2–4. Our numerical codes give the same results as the analytic expressions of Eq. (11). This provides a check that our algebra is correct. We note that $2n_1n_2/(n_1+n_2)^2\leqslant\frac{1}{2}$ and that the maximum value of $P_3(t)$ occurs for $n_1=n_2$, where $P_{3max}=\frac{1}{2}$, consistent with Fig. 2. This corresponds to $\alpha=0$ so that direct transitions from state one to state two are forbidden. Transfer to state two occurs via the intermediate state three. Transfer from state one to state two and back is complete, and occurs periodically. In general $\alpha=0$ occurs when $n_1=n_2=3n_{odd}$, where n_{odd} is any odd integer. This appears to give the simplest condition that allows complete population transfer. In this case the action area is $A(t_0)=n_{odd}\pi/\sqrt{2}$.

Calculations for two other values of α that allow complete transfer to state two are shown in Figs. 3 and 4. We see

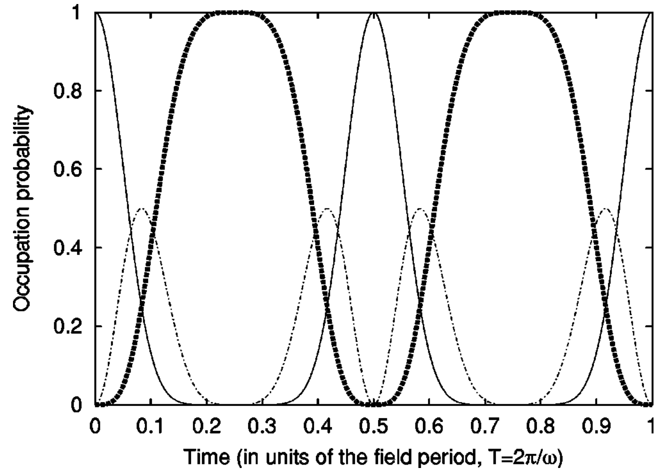

FIG. 2. Occupation probabilities as a function of time. The thick dashed line denotes $P_2(t)$, the solid line denotes $P_1(t)$, and the thin dash-dotted line denotes $P_3(t)$. In this figure we use the allowed values, $\alpha=V_{12}/V_{23}=0$ and $A(t_0)=\int_0^{t_0}V(t')dt'=2.221$, corresponding to $n_1=3$ and $n_2=3$ ($n_o=n_o'=1$) in Eq. (10). Complete transfer to state two from state one occurs at $t=t_0=T/4$ and again at odd multiples of t_0.

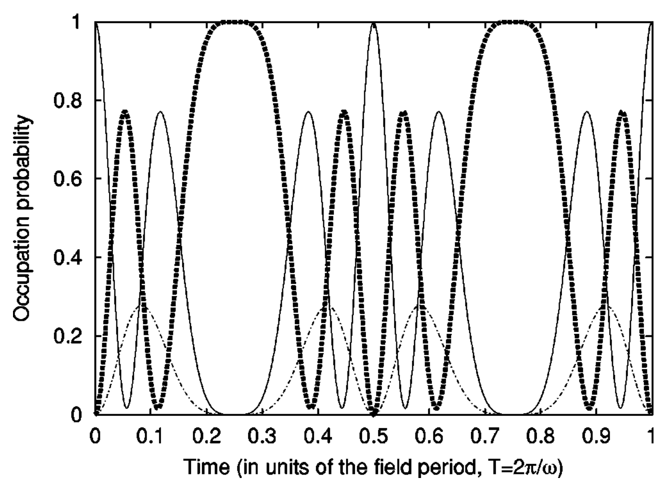

FIG. 3. Occupation probabilities as a function of time. The thick dashed line denotes $P_2(t)$, the solid line denotes $P_1(t)$, and the thin dash-dotted line denotes $P_3(t)$. In this figure $\alpha=-2.530$ and $A(t_0)=1.656$, corresponding to $n_1=1$ and $n_2=5$ ($n_o=-1$, $n_o'=3$) in Eq. (10).

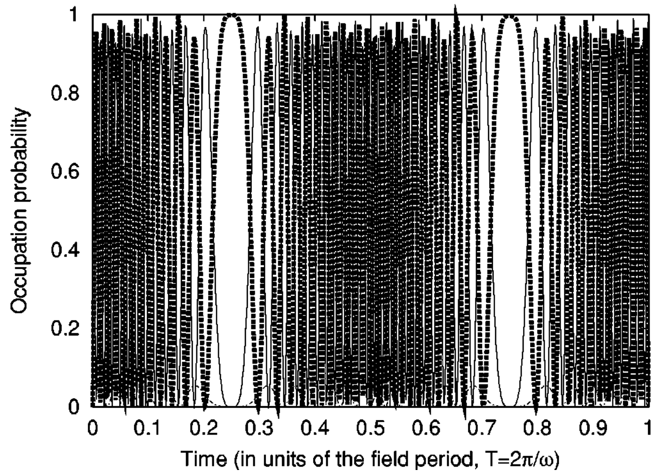

FIG. 4. Occupation probabilities as a function of time. The thick dashed line shows $P_2(t)$, the solid line shows $P_1(t)$, and the thin dash-dotted line shows $P_3(t)$. Here $\alpha=8.128$ and $A(t_0)=4.381$, corresponding to $n_1=35$ and $n_2=1$ ($n_o=23$, $n_o'=-11$) in Eq. (10). Since these values are allowed, the electron population is completely transferred to state two from state one.

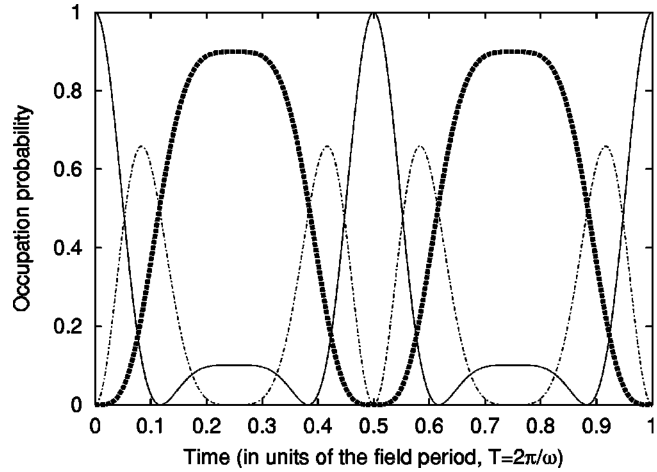

FIG. 5. Occupation probabilities as a function of time in atomic lithium. Solid line, initial state $4p_0$; thin dash-dotted line, intermediate state $4d_0$; thick dashed line, target state $4f_0$.

that complete transfer occurs twice in one period of the oscillating field but that the frequency of the "side bands" increases as αA increases. We note that $\alpha=\pm\sqrt{2/(n_1 n_2)}\,(n_1-n_2)$ becomes either large ($n_1 \gg n_2$, or vice versa), or small ($n_1 \sim n_2$) as $n_1 n_2$ increases, while $A(t_0)$ increases as $\sqrt{n_1 n_2/2}$. When complete population transfer occurs, the population lingers in state two, as seen in Figs. 2–4.

III. RESULTS AND DISCUSSION

In this paper we have considered transitions amplitudes and probabilities in a three-state atom with degenerate states. In realistic atomic systems, energy states are seldom, if ever, exactly degenerate. Also states outside three-state manifold usually exist. However, our method can be used for atoms with a certain structure of energy states, namely, for atoms that have three nearly degenerate states separated from the other states by a relatively big energy gap. One example of such system is a hydrogen atom (transitions between $3s$, $3p_0$, and $3d_0$ states, see Ref. [29]). Another example is atomic lithium ($4p_0$, $4d_0$, and $4f_0$ states). In both cases the energy structure of the states imposes restrictions on the frequency of the external field, $\omega_{min}<\omega<\omega_{max}$. Here $\hbar\omega_{min}$ is the energy splitting of the nearly degenerate states, and $\hbar\omega_{max}$ is the energy difference between the three-state manifold and the closest state in energy outside the manifold. In both cases conventional $E1$ dipole selection rules apply for linearly polarized light. Calculations are shown for lithium in Fig. 5, based on oscillator strengths [30] that give $\alpha=1.39$ (with $\beta=0$ due to the dipole selection rule). Three closely spaced energy levels also occur in the ground states of atomic carbon, oxygen, and silicon. These atoms all have three states with different values of a total angular momentum. Magnetic dipole or electric quadrupole transitions can be used for transitions that couple $J=0,1,2$. For example, the $2s^2 2p^{2\,3}P$ term of ${}^{12}_{6}C$ has three levels ($J=0,1,2$) separated by less than 0.006 eV, while the closest adjacent level ($2s^2 2p^{2\,1}D$ has an energy 1.26 eV. This corresponds to a ratio $\omega_{max}/\omega_{min}\approx 234$, indicating that the criteria for application of our method is well satisfied. The corresponding ratios for atomic oxygen and silicon are 70 and 28, respectively. More generally three degenerate levels of $J=1$ atomic states may also be controlled using magnetic dipole transitions.

In lithium energy difference between $4p_0$, $4d_0$, and $4f_0$ states is less than 0.02 eV, while the closest adjacent $4s$ state is 0.18 eV apart. Therefore, one can use photons with energies in the range 0.02 eV$<\hbar\omega<$0.18 eV. When the states are nondegenerate $\omega_{ij}=(E_i-E_j)/\hbar\neq 0$, and the population transfer is incomplete. Numerical calculations indicate that this nondissipative population leakage varies as $(\omega_{ij}/\omega)^2$. For $\hbar\omega=0.1$ eV this factor is 0.04 for lithium. The choice of the field frequency ω involves a trade-off between the duration of time the population remains in state two and the population leakage. The higher the frequency ω the smaller the population leakage, but the shorter the duration time T_s that the population remains in state two. This effect of population leakage is similar as that for a two-state atom [28]. Another factor that prevents complete transfer is the relative strength of the matrix elements, α and β. For the transitions under consideration their values ($\alpha=0,\beta=1.39$) differ from the one that allows the complete transfer ($\alpha=0,\beta=1$) corresponding to Fig. 2. As a result, the occupation probability for the target state never exceeds 90% (cf. Fig. 5).

The duration of time spent in the transferred state can be controlled by adjusting the time dependence of the external field. If the external interaction varies harmonically, $V(t)=V_0\cos(\omega t)$, then the first line of Eq. (10) imposes a constraint between V_0 and ω, namely, $V_0/\omega=\pm\sqrt{n_1 n_2/2}\,\pi/3$. For a harmonic interaction the duration of the time T_s that the population remains in state two varies inversely with ω; the higher the frequency ω the smaller the time the population remains in state two. The condition $\alpha=0$ corresponds to a dipole selection rule. Transfer from state one to state two (which is the dipole forbidden transition) is complete at time

t_0 if the two allowed transition matrix elements, V_{23} and V_{13}, are equal in absolute magnitude and $A(t_0)=n_{odd}\pi/2$. In some atomic systems state two could decay, with a lifetime T_d to another state outside the degenerate manifold and be lost. Such loss can be controlled by adjusting ω.

Additional control [28] may be achieved by changing the shape of $V(t)$. This can be used to control how long the population remains near unity in state two, for example. An interesting example is the case of an ideal, sudden ''kick'' produced by $V(t)=A_0\delta(t-t_0)$. If $A_0\rightarrow n_{odd}\pi/\sqrt{2}$, for example, then the mere presence of state three allows transfer from state one to state two without any direct transfer from state one to state two. With this ideal kick, state two is unoccupied before $t=t_0$, and fully occupied after $t=t_0$. This is a switch. One may apply a series of switches by taking $V(t)=A_0[\delta(t-t_1)+\delta(t-t_2)+\cdots+\delta(t-t_k)]$. Alternatively one may switch to different states by adjusting the matrix elements so that after each step $V_{12}(t)$ is interchanged with $V_{1'2}(t)$, where state $1'$ is now the state occupied before the next kick is applied. Two practical limitations on this ideal model are the impossibility of producing a signal that varies as $\delta(t-t_0)$, and the existence of an infinitely wide spectrum of high-frequency components with frequencies $\omega>\omega_{max}$ in the Fourier spectrum of $\delta(t-t_0)$. Fortunately these two difficulties can both be addressed by using kicks of finite width in time. In some cases it may be possible to design a kick so that its duration is short compared to any other changes in the system, so that a finite kick may be sensibly represented by $\delta(t-t_0)$.

We note that the method described here is generally applicable to a degenerate n-state system [31] in a case where many of the matrix elements of the external interaction are the same, since such a degenerate n-state system can be reduced to a mathematically equivalent three-state system. However, the conditions placed on the matrix elements are not often met in most atomic systems. Finally we remark that use of degeneracy, $\Delta E\rightarrow 0$, imposes $\Delta t\rightarrow\infty$. This removes time sequencing in intermediate steps of the reaction process. That is, any physical effect due to time sequencing of intermediate interactions is lost in the limit of degeneracy.

IV. SUMMARY

We have shown that in a three-state atom with degenerate energies, electron population is completely transferred via an external interaction at a designated time t_0 from an initially occupied state (state one) to a designated initially unoccupied state (state two) under two conditions. The first condition for complete transfer is that the ratio of the matrix elements of the external interaction, $V_{ij}(t)$, satisfy $V_{12}(t)/V_{23}(t)=\alpha=\pm\sqrt{2/(n_1n_2)}\,(n_1-n_2)$, and $V_{13}(t)/V_{23}(t)=\beta=\pm 1$. The second condition is that at $t=t_0$ the action area of $V(t)$ satisfy $A(t_0)=\int_0^{t_0}V(t')dt'=\pm\sqrt{n_1n_2/2}\,\pi/3$, where we have set $V_{23}(t)=V(t)$. Here n_1 and n_2 are integers such that $n_1=2n_o+n_o'$ and $n_2=n_o+2n_o'$, where n_o and n_o' are any odd integers. The duration of time the transferred population remains in state two can be controlled either by varying the frequency ω of the external potential or by varying the shape of $V(t)$.

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APPENDIX A: METHOD TO FIND CONDITIONS FOR COMPLETE POPULATION TRANSFER

Here we show how to find the conditions on the V_{jk} such that the electron population is completely transferred from its initial state $i=1$ to a different final state, $i=2$ or 3. A convenient way to begin is to take $dP_k(t)/dt=0$ at $t=t_0$. This gives both maxima and minima for P_k. Using Eq. (8) this yields

$$\sin((z_i-z_j)A(t_0))=0,$$

$$(z_i-z_j)A(t_0)=m_{ij}\pi \quad (m_{ij}\neq 0) \quad (i,j=1,2,3). \tag{A1}$$

Using $\cos((z_i-z_j)A(t_0))=\cos(m_{ij}\pi)=(-1)^{m_{ij}}$, one has

$$\begin{aligned}P_1=|a_1|^2=&\frac{1}{\Delta^2}[(x_2y_3-x_3y_2)^2+(x_3y_1-x_1y_3)^2+(x_1y_2\\&-x_2y_1)^2+2(-1)^{m_{12}}(x_2y_3-x_3y_2)(x_3y_1-x_1y_3)\\&+2(-1)^{m_{13}}(x_2y_3-x_3y_2)(x_1y_2-x_2y_1)\\&+2(-1)^{m_{23}}(x_3y_1-x_1y_3)(x_1y_2-x_2y_1)]\\=&\frac{1}{\Delta^2}[(-1)^{k_1}(x_2y_3-x_3y_2)+(-1)^{k_2}(x_3y_1-x_1y_3)\\&+(-1)^{k_3}(x_1y_2-x_2y_1)]^2,\end{aligned} \tag{A2}$$

where $k_1+k_2=m_{12}$, $k_1+k_3=m_{13}$ and $k_2+k_3=m_{23}$. Now,

$$\begin{aligned}k\equiv k_1&=\frac{1}{2}(m_{12}+m_{13}-m_{23})\\&=\frac{1}{2}\left(\frac{A(t_0)}{\pi}(z_1-z_2)+\frac{A(t_0)}{\pi}(z_1-z_3)-\frac{A(t_0)}{\pi}(z_2-z_3)\right)\\&=\frac{A(t_0)}{\pi}(z_1-z_2),\end{aligned}$$

$$\begin{aligned}k_2&=\frac{1}{2}(m_{12}+m_{23}-m_{13})\\&=\frac{1}{2}\left(\frac{A(t_0)}{\pi}(z_1-z_2)+\frac{A(t_0)}{\pi}(z_2-z_3)-\frac{A(t_0)}{\pi}(z_1-z_3)\right)\\&=0,\end{aligned}$$

$$k' \equiv k_3 = \frac{1}{2}(m_{13}+m_{23}-m_{12})$$
$$= \frac{1}{2}\left(\frac{A(t_0)}{\pi}(z_1-z_3)+\frac{A(t_0)}{\pi}(z_2-z_3)-\frac{A(t_0)}{\pi}(z_1-z_2)\right)$$
$$= \frac{A(t_0)}{\pi}(z_2-z_3). \tag{A3}$$

This yields extrema for P_k at $t=t_0$, namely,

$$P_1 = \frac{1}{\Delta^2}[(-1)^k(x_2y_3-x_3y_2)+(x_3y_1-x_1y_3) + (-1)^{k'}(x_1y_2-x_2y_1)]^2,$$

$$P_2 = \frac{1}{\Delta^2}[(-1)^k(y_2-y_3)+(y_3-y_1)+(-1)^{k'}(y_1-y_2)]^2,$$

$$P_3 = \frac{1}{\Delta^2}[(-1)^k(x_3-x_2)+(x_1-x_3)+(-1)^{k'}(x_2-x_1)]^2, \tag{A4}$$

where k and k' are nonzero integers that are *not both even*.

1. Conditions for complete transfer

We now seek conditions such that at $t=t_0$, $P_1=0$, $P_2=1$, and $P_3=0$. There are three sets, $\{k,k'\}$, that satisfy these conditions, namely, (i) k is even and k' is odd, (ii) k and k' are both odd, and (iii) k is odd and k' is even.

Case (i). k is even and k' is odd. Then,

$$P_1 = \frac{1}{\Delta^2}[(x_2y_3-x_3y_2)+(x_3y_1-x_1y_3)-(x_1y_2-x_2y_1)]^2,$$

$$P_2 = \frac{1}{\Delta^2}[(y_2-y_3)+(y_3-y_1)-(y_1-y_2)]^2 = \frac{4}{\Delta^2}(y_2-y_1)^2,$$

$$P_3 = \frac{1}{\Delta^2}[(x_3-x_2)+(x_1-x_3)-(x_2-x_1)]^2 = \frac{4}{\Delta^2}(x_1-x_2)^2. \tag{A5}$$

From the condition that $P_2=1$ one has $[2(y_2-y_1)/\Delta]^2=1$, where $y_1 \neq y_2$, so that $\Delta = \pm 2(y_2-y_1)$. The condition that $P_3=0$ gives $[2(x_1-x_2)/\Delta]^2=0$, which yields $x_1=x_2=x$. Using this result one then obtains $P_1=[(x+x_3)(y_1-y_2)/\Delta]^2$. Requiring $P_1=0$, one has $x_3=-x$. Now, using $x_1=x_2=-x_3=x$, we have from below Eq. (7), $\Delta=2x(y_2-y_1)$. Finally since $\Delta=\pm 2(y_2-y_1)$, one has $x_1=x_2=-x_3=x=\pm 1$.

Case (ii). k and k' are both odd. Then,

$$P_1 = \frac{1}{\Delta^2}[-(x_2y_3-x_3y_2)+(x_3y_1-x_1y_3)-(x_1y_2-x_2y_1)]^2,$$

$$P_2 = \frac{1}{\Delta^2}[-(y_2-y_3)+(y_3-y_1)-(y_1-y_2)]^2$$
$$= \frac{4}{\Delta^2}(y_3-y_1)^2,$$

$$P_3 = \frac{1}{\Delta^2}[-(x_3-x_2)+(x_1-x_3)-(x_2-x_1)]^2$$
$$= \frac{4}{\Delta^2}(x_1-x_3)^2. \tag{A6}$$

From the condition that $P_2=1$ one has $[2(y_3-y_1)/\Delta]^2=1$, where $y_1 \neq y_3$, so that $\Delta=\pm 2(y_3-y_1)$. The condition that $P_3=0$ gives $[2(x_1-x_3)/\Delta]^2=0$, which yields $x_1=x_3=x$. Then one obtains $P_1=[(x+x_2)(y_1-y_3)/\Delta]^2$. When $P_1=0$ one has $x_2=-x$. Now, using $x_1=-x_2=x_3=x$, we get from below Eq. (7), $\Delta=2x(y_1-y_3)$. Since $\Delta=\pm 2(y_3-y_1)$, one has $x_1=-x_2=x_3=x=\mp 1$.

Case (iii). k is odd and k' is even. This is similar to case (i). One obtains, $\Delta=2x(y_3-y_2)$ and $-x_1=x_2=x_3=x=\pm 1$.

Complete population transfer, i.e., $P_2=1$, requires that $x=\pm 1$, so that the relative interaction strengths α and β must satisfy

$$\alpha = \frac{2}{y} - y \pm \frac{\epsilon_2-\epsilon_1}{y^2} \pm (\epsilon_3-\epsilon_2),$$

$$\beta = \pm 1 + \frac{\epsilon_2-\epsilon_1}{y}. \tag{A7}$$

Next we use these results to find the conditions on V_{ij} required for complete population transfer to occur.

2. Quantum numbers for $A(t_0)$ and α

We now note that relatively simple conditions occur when $\epsilon_1=\epsilon_2=\epsilon_3=\epsilon$, namely, $\alpha=(2/y)-y$ and $\beta=\pm 1$. In this case Eq. (A7) reduces from a cubic to an easier quadratic equation in y. This quadratic equation corresponds to three roots of the more general cubic equation, namely,

$$\{y_j\} = \{y_+, y_-, 0\} = \left\{\frac{-\alpha+\sqrt{\alpha^2+8}}{2}, \frac{-\alpha-\sqrt{\alpha^2+8}}{2}, 0\right\}. \tag{A8}$$

Now for case (i) when k is even and k' is odd, one has from above, $x=\{\pm 1, \pm 1, \mp 1\}$ and $y=\{y_+, y_-, 0\}$. From Eq. (A8), $z=\{\mp y_-, \mp y_+, \mp \alpha\}$. Then, with $E=A(t_0)/\pi$, the useful integers k and k' are: $k=A(t_0)/\pi(z_1-z_2)=\pm E\sqrt{\alpha^2+8}$, and $k'=A(t_0)/\pi(z_2-z_3)=\pm E/2(3\alpha-\sqrt{\alpha^2+8})$. Eliminating α, one obtains $(2k+k')(k-k')=18E^2=n_1n_2$. For case (ii) when k and k' are both odd, $x=\{\pm 1, \mp 1, \pm 1\}$ and $y=\{y_+, 0, y_-\}$. and $z=\{\mp y_-, \mp \alpha, \mp y_+\}$. Then, $k=A(t_0)/\pi(z_1-z_2)=\pm E/2(3\alpha+\sqrt{\alpha^2+8})$, and $k'=A(t_0)/\pi(z_2-z_3)=\mp E/2(3\alpha-\sqrt{\alpha^2+8})$. One may now verify that $(2k+k')(k+2k')=18E^2=n_1n_2$. For case (iii) when k is odd and k' is even, $x=\{\mp 1,\pm 1,\pm 1\}$ and $y=\{0,y_+,y_-\}$, and $z=\{\mp\alpha,\mp y_-,\mp y_+\}$. Then, $k=A(t_0)/\pi(z_1-z_2)=\mp E/2(3\alpha+\sqrt{\alpha^2+8})$, and $k'=A(t_0)/\pi(z_2-z_3)=\pm E\sqrt{\alpha^2+8})$. One may again verify that $(k'-k)(2k'+k)=18E^2=n_1n_2$.

One readily finds in case (i), where k is even and k' is odd, that $k=\frac{1}{3}(n_1+n_2)$ and $k'=\frac{1}{3}(n_1-2n_2)$. In case (ii), where k and k' are both odd, one has $k=\frac{1}{3}(2n_1-n_2)$ and $k'=\frac{1}{3}(2n_2-n_1)$. In case (iii) $k=\frac{1}{3}(n_2-2n_1)$ and $k'=\frac{1}{3}(n_1+n_2)$. In all three cases one readily obtains the same equation for the allowed values of $A(t_0)$ and α, namely,

$$A(t_0)=\pm\sqrt{\frac{n_1n_2}{2}}\frac{\pi}{3},$$

$$\alpha=\pm\sqrt{\frac{2}{n_1n_2}}(n_1-n_2). \tag{A9}$$

It is remarkable that all three cases give the same formula for complete population transfer. The top equation corresponds to a constraint on the value of the action, $\int_0^t V(t')dt'$, i.e., the "area" of the external interaction, $V(t)$, or the phase shift caused by the external interaction. The bottom equation above gives the relative values of the V_{ij}.

APPENDIX B: GENERAL SOLUTIONS FOR THE DEGENERATE THREE-STATE ATOM

In the main text a solution for the degenerate three-state atom was found in the case when $\epsilon_j=0$. We now choose $\epsilon_j^{-1}V_{jj}(t)=V(t)=V_{23}(t)=\alpha^{-1}V_{12}(t)=\beta^{-1}V_{13}(t)$. Then Eq. (2) becomes

$$i\dot{a}_1(t)=\epsilon_1V(t)a_1(t)+\alpha V(t)a_2(t)+\beta V(t)a_3(t),$$

$$i\dot{a}_2(t)=\alpha V(t)a_1(t)+\epsilon_2V(t)a_2(t)+V(t)a_3(t),$$

$$i\dot{a}_3(t)=\beta V(t)a_1(t)+V(t)a_2(t)+\epsilon_3V(t)a_3(t). \tag{B1}$$

Consider a linear superposition $c(t)=a_1(t)+xa_2(t)+ya_3(t)$, where x and y are some time-independent coefficients that we next determine. Then,

$$\begin{aligned}i\dot{c}(t)&=i\dot{a}_1(t)+xi\dot{a}_2(t)+yi\dot{a}_3(t)\\&=\epsilon_1V(t)a_1(t)+\alpha V(t)a_2(t)+\beta V(t)a_3(t)\\&\quad+x[\alpha V(t)a_1(t)+\epsilon_2V(t)a_2(t)+V(t)a_3(t)]\\&\quad+y[\beta V(t)a_1(t)+V(t)a_2(t)+\epsilon_3V(t)a_3(t)].\end{aligned} \tag{B2}$$

Set $z=\epsilon_1+\alpha x+\beta y$. Then, $i\dot{c}(t)=zV(t)[a_1(t)+(\alpha+\epsilon_2x+y)/(\epsilon_1+\alpha x+\beta y)a_2(t)+(\beta+x+\epsilon_3y)/(\epsilon_1+\alpha x+\beta y)a_3(t)]$. We require that $i\dot{c}(t)=zV(t)c(t)$. This holds if and only if $z=\epsilon_1+\alpha x+\beta y$, $x=(\alpha+\epsilon_2x+y)/(\epsilon_1+\alpha x+\beta y)$ and $y=(\beta+x+\epsilon_3y)/(\epsilon_1+\alpha x+\beta y)$. After some algebra this leads to the useful cubic equation

$$\begin{aligned}&[(\beta^2-\alpha^2)+\alpha\beta(\epsilon_2-\epsilon_3)]y^3+[\alpha(2-\alpha^2-\beta^2)+\beta(2\epsilon_1-\epsilon_2\\&\quad-\epsilon_3)+\alpha(\epsilon_1-\epsilon_3)(\epsilon_2-\epsilon_3)]y^2+[(2\alpha^2-\beta^2-1)\\&\quad+\alpha\beta(2\epsilon_3-\epsilon_1-\epsilon_2)+(\epsilon_1-\epsilon_2)(\epsilon_1-\epsilon_3)]y\\&\quad+[\alpha(\beta^2-1)-\beta(\epsilon_1-\epsilon_2)]=0.\end{aligned} \tag{B3}$$

This determines three sets of eigenvalues, $\{x_j\}$, $\{y_j\}$, and $\{z_j\}$, and three eigenfunctions, $c_j(t)=e^{-iz_jA(t)}$, where $A(t)=\int_0^t V(t')dt'$.

When $\epsilon_j\neq 0$ complete population transfer can also occur. Using $\epsilon_j=\epsilon\neq 0$ adds nothing new, since it produces only an overall phase $e^{i\epsilon t}$. A slightly more general, but less elegant, solution for the degenerate three-state equations is found when $\epsilon_1=\epsilon_2\neq\epsilon_3$. Again in this case $\alpha=(2/y)-y\pm(\epsilon_3-\epsilon_1)$ and $\beta=\pm 1$. The quadratic equation for y now becomes $y^2+[(\alpha(1-\alpha^2)+\alpha(\epsilon_1-\epsilon_3)^2\pm(\epsilon_1-\epsilon_3))/(1-\alpha^2\pm\alpha(\epsilon_1-\epsilon_3))]y-2=0$. Taking $\tilde{\alpha}=(\alpha(1-\alpha^2)+\alpha(\epsilon_1-\epsilon_3)^2\pm(\epsilon_1-\epsilon_3))/(1-\alpha^2\pm\alpha(\epsilon_1-\epsilon_3)=\alpha\pm(\epsilon_1-\epsilon_3))$ the algebra is the same as that one above, except that $\alpha\to\tilde{\alpha}$. This includes an additional parameter, $(\epsilon_1-\epsilon_3)$, gives simple allowed values of $\tilde{\alpha}$, and imposes a constraint between α and $(\epsilon_1-\epsilon_3)$. More complex conditions for complete population transfer might exist when $\epsilon_1\neq\epsilon_2$, in which case Eq. (B3) is cubic in y. Finally it might be possible that solutions exist when the matrix elements $V_{ij}(t)$ have different time dependences, i.e., when α and β depend on t.

Conditions for complete transfer into state three at $t=t_0$ may be generally found similarly by interchange of the indices 2 and 3, with corresponding interchange of α with β and x with y.

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