Analytic Solution for the Nondegenerate Quantum Control Problem

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We present an analytic solution for the nondegenerate quantum control problem, i.e., the transfer of a deliberate amount of population, 0%–100%, between arbitrary initial $|\Psi(t)\rangle$ and final $|\Psi'(t)\rangle$ states, which can be expanded in terms of nondegenerate energy eigenstates $|k\rangle$. The solution constitutes a robust two-photon multicomponent adiabatic passage, via an intermediate eigenstate $|0\rangle$, which relies on three types of "null states."

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The "quantum control" problem consists of finding ways of causing a complete transfer of population from an arbitrary initial state to a desired "target" state, under the guidance of laser pulses. Its solution has been the central theme in the fields of coherent control [1] and optimal control [2,3]. Much attention was also devoted to the more restricted problem of "nondegenerate quantum control," where population is transferred between superpositions of nondegenerate energy eigenstates. The reason is that one can find such nondegenerate spectra in many atomic and molecular systems [4].

Although existence theorems have been derived [5,6], and analytic solutions are known in some weak field [1] and strong field regimes, e.g., the stimulated Raman adiabatic passage method [7,8], so far no analytic solution has been presented for complete transitions between *arbitrary superpositions* of energy eigenstates [9]. Optimal control practitioners have treated this general problem by using brute-force search techniques [2,6] for shaped light pulses [10,11], which perform the desired task. These involve a complex iterative (nonlinear) solution of the matter + radiation Schrödinger equation. Recently [12], we have presented an analytic solution to a yet more restricted problem of transferring a *single* energy eigenstate $|1\rangle$ to a superposition of other energy eigenstates $|\Psi'\rangle = \sum_k c_k e^{-i\omega_k t}|k\rangle$. We have shown that complete population transfer to the target state can be achieved by performing an adiabatic passage via an intermediate eigenstate $|0\rangle$, induced by two laser pulses whose shapes have been derived analytically. We have termed this method "shaped adiabatic passage" (SAP). Related to the outlined general problem is the idea of a *multiple* complete population transfer, studied so far through a chain of single levels [13].

In this work, we solve the nondegenerate quantum control problem by showing that a technique superior to SAP can be applied to the transfer of population from an *initial superposition state*. The process which follows the sequence of events, $|\Psi\rangle = \sum_k c_k e^{-i\omega_k t}|k\rangle \rightarrow |0\rangle \rightarrow |\Psi'\rangle = \sum_{k'} c'_{k'} e^{-i\omega_{k'} t}|k'\rangle$, can be deliberately *repeated* to produce other desired superposition states $|\Psi''\rangle, |\Psi'''\rangle \ldots$. Moreover, three types of "null states" allow us to control the degree of population transfer, from 0% to 100%.

Figure 1 illustrates the method with the counterintuitive pulse ordering [7]. In this scheme, the "dump" pulse with Rabi frequencies $\Omega_{0,k'}$, coupling empty $|0\rangle$ and $|k'\rangle$ ($k' = n+1, \ldots, n+m$) states, *precedes* (while partially overlapping) the "pump" pulse with Rabi frequencies $\Omega_{k,0}$, coupling the populated $|k\rangle$ ($k = 1, \ldots, n$) states to the $|0\rangle$ state. As shown below, for certain choices of the $\Omega_{k,0}$ and $\Omega_{0,k'}$ frequencies, an adiabatic and robust transfer of a *chosen* amount of population takes place between the $|\Psi\rangle$ and $|\Psi'\rangle$ wave packets.

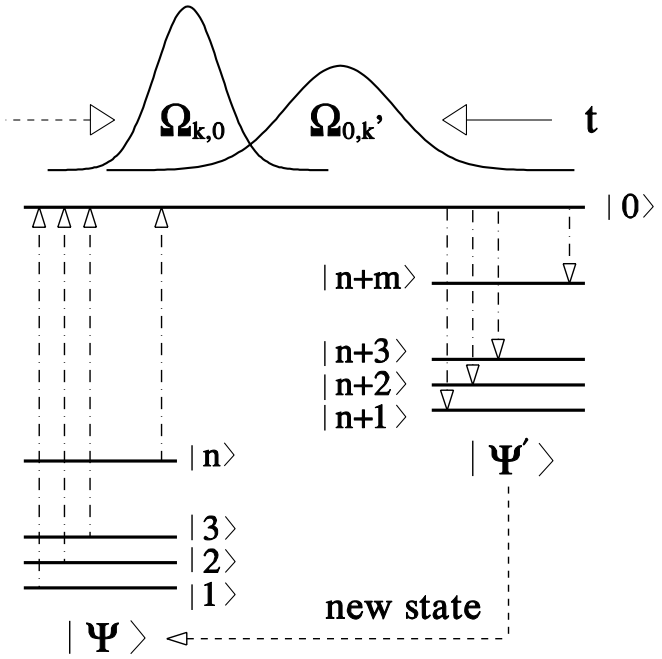

FIG. 1. A multiple transfer scheme between the states $|\Psi\rangle \rightarrow |0\rangle \rightarrow |\Psi'\rangle \rightarrow |0\rangle \rightarrow |\Psi''\rangle \ldots$. The first (dump) laser pulse, with Rabi frequencies $\Omega_{0,k'}(t)$, couples all the $|k'\rangle$ final (empty) states to state $|0\rangle$, while the second (pump) pulse, with Rabi frequency $\Omega_{k,0}(t)$, couples state $|0\rangle$ to the initial (populated) states $|k\rangle$.

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In order to explain the effect, we consider the action of a pulse, represented by a multimode electric field,

$$\mathcal{E}(t) = \mathcal{R}_e \sum_{k=1}^{n+m} \mathcal{E}_{0,k}(t)\, e^{-i\omega_{0,k}t}, \tag{1}$$

on a superposition of $|k\rangle$ levels with eigenenergies ω_k. In the above $\mathcal{E}_{0,k}(t)$ are the (slowly varying) amplitudes of the $\omega_{0,k} \equiv \omega_0 - \omega_k$ modes, which resonantly couple the $|0\rangle$ and $|k\rangle$ levels. We construct a sequence of two pulses, by choosing *one* time dependence ($0 < f_D(t) < 1$) for the $\mathcal{E}_{0,k}(t)$ amplitudes of the dump process, which is connecting the $|k\rangle = |n+1\rangle, |n+2\rangle, \ldots, |n+m\rangle$ states to the $|0\rangle$ state, and *another* time (delayed) dependence ($0 < f_P(t) < 1$) for the $\mathcal{E}_{0,k}(t)$ amplitudes of the pump process, connecting the $|0\rangle$ state to the $|k\rangle = |1\rangle, |2\rangle, \ldots, |n\rangle$ states. The intensity and phase of each $\mathcal{E}_{0,k}(t)$ amplitude is adjusted to yield the desired transfer, as discussed below.

We can write the matter + radiation Hamiltonian in the rotating waves approximation, neglecting off-resonance terms, in atomic units ($\hbar = 1$), as

$$H = \sum_{k=0}^{n+m} \omega_k |k\rangle\langle k| + \sum_{k=1}^{n+m} [\Omega_{0,k}(t) e^{-i\omega_{0,k}t} |0\rangle\langle k| + \text{H.c.}].$$

Here, $\Omega_{0,k}(t) \equiv \mu_{0,k}\, \mathcal{E}_{0,k}(t) \equiv \mathcal{O}_{0,k}\, f_{D(P)}(t)$ are the complex Rabi frequencies, with $\mu_{0,k}$ being the electric-dipole matrix elements between the $|0\rangle$ and $|k\rangle$ states.

The system evolution is described by the wave function $|\Psi\rangle = \sum_{k=0}^{n+m} c_k(t)\, e^{-i\omega_k t} |k\rangle$. The vector $\mathbf{c}(t)$ of the $c_k(t)$ coefficients, $\mathbf{c}(t) = (c_0, c_1, \ldots, c_n, c_{n+1}, \ldots, c_{n+m})$, is a solution of the matrix Schrödinger equation

$$\dot{\mathbf{c}}^{\mathsf{T}}(t) = -i\mathsf{H}(t)\, \mathbf{c}^{\mathsf{T}}(t), \tag{2}$$

where T designates the matrix transpose, and the effective time-dependent Hamiltonian is

$$\mathsf{H}(t) = \begin{bmatrix} 0 & \Omega_{0,1} & \cdots & \Omega_{0,n+m} \\ \Omega_{1,0} & 0 & \cdots & 0 \\ \cdots & \cdots & \cdots & \cdots \\ \Omega_{n+m,0} & 0 & \cdots & 0 \end{bmatrix}.$$

In the counterintuitive scheme, the pump pulse with the $\mathcal{E}_{1,0}, \ldots, \mathcal{E}_{n,0}$ components follows the dump pulse with the $\mathcal{E}_{n+1,0}, \ldots, \mathcal{E}_{n+m,0}$ components.

Of the $n+m+1$ eigenvalues of $\mathsf{H}(t)$, $n+m-1$ are zero, $\lambda_1 = \lambda_2 = \ldots = \lambda_{n+m-1} = 0$, and two are nonzero, $\lambda_{n+m}(t) = -\lambda_{n+m+1}(t) = (\sum_{k=1}^{n+m} |\Omega_{0,k}(t)|^2)^{1/2}$. The crucial zero eigenvalues correspond to *three* types of null states, which we term as the "initial null states" (INS), the "mixed null states" (MNS), and the "final null states" (FNS). Redefining the basis vectors as $e^{-i\omega_j t}|j\rangle \to |j\rangle$, the three types of null states are

$$\begin{aligned} |D^I_{kk'}\rangle &= \Omega_{0,k'}|k\rangle - \Omega_{0,k}|k'\rangle, \\ |D^M_{kl}\rangle &= \Omega_{0,l}|k\rangle - \Omega_{0,k}|l\rangle, \\ |D^F_{ll'}\rangle &= \Omega_{0,l'}|l\rangle - \Omega_{0,l}|l'\rangle, \\ &(k \neq k' = 1, \ldots, n; l \neq l' = n+1, \ldots, n+m). \end{aligned} \tag{3}$$

In this construction we can produce $n(n-1)/2$, nm, and $m(m-1)/2$ INS, MNS, and FNS, respectively, where only $n+m-1$ states are linearly independent.

We first examine the MNS states and show that we can combine them to obtain a state that correlates (in the counterintuitive pulse ordering) at $t = 0$ with the initial state $|\Psi\rangle = \sum_k c_k^0 |k\rangle$, and at $t = t_{\text{end}}$ with the final state $|\Psi'\rangle = \sum_l c_l^e |l\rangle$. The particular combination that satisfies these asymptotic conditions is

$$\begin{aligned} |D^M\rangle &= \sum_{k,l} t_{kl} |D^M_{kl}\rangle \\ &= \sum_{k=1}^{n} |k\rangle \sum_{l=n+1}^{n+m} t_{kl}\Omega_{0,l} - \sum_{l=n+1}^{n+m} |l\rangle \sum_{k=1}^{n} t_{kl}\Omega_{0,k}, \end{aligned} \tag{4}$$

where the t_{kl} coefficients are chosen, so that

$$\sum_{l=n+1}^{n+m} t_{kl}\Omega_{0,l} \propto c_k^0, \qquad \sum_{k=1}^{n} t_{kl}\Omega_{0,k} \propto c_l^e. \tag{5}$$

Equations (5) can be satisfied by choosing $[\Omega_{k,0}(t) \equiv \mathcal{O}_{k,0}\, f_{D(P)}(t)]$

$$t_{kl} = \mathcal{O}_{k,0}\mathcal{O}_{l,0}, \quad \text{and} \quad \mathcal{O}_{k,0} = C c_k^0, \qquad \mathcal{O}_{l,0} = C' c_l^e, \tag{6}$$

where C, C' are arbitrary complex numbers. The only limitation on their choice is that the (slowly varying) Rabi frequencies should be strong enough to guarantee the adiabaticity of the transfer process. Since the MNS of Eq. (4) is degenerate with other null states, adiabaticity could be easily broken by their possible coupling. But when the chosen MNS is orthogonalized to the rest null states, these couplings become zero, similarly as in SAP [12], so the transfer $|\Psi\rangle \to |\Psi'\rangle$ is robust.

We can directly apply Eqs. (4)–(6) in practical problems. Suppose that we want to switch between vibrational wave packets, $|\Psi(0)\rangle = \sum_{v=1}^{n} c_v^0 |X^1\Sigma_g^+, v\rangle$ and $|\Psi'(t_{\text{end}})\rangle = \sum_{v=n+1}^{n+m} c_v^e |X^1\Sigma_g^+, v\rangle$, sitting on the *ground* electronic state $X^1\Sigma_g^+$ of the Na_2 molecule [12], and use the *excited* state $|A^1\Sigma_u^+, v' = 0\rangle$ as the intermediate state $|0\rangle$. The transition-dipole matrix elements are $\mu_{0,v} \equiv \langle A^1\Sigma_u^+, v' = 0|\hat{\boldsymbol{\epsilon}} \cdot \boldsymbol{\mu}|X^1\Sigma_g^+, v\rangle \approx \bar{\mu}\langle 0|v\rangle$, where $\bar{\mu} \approx 7$ D is the "average" transition-dipole moment and $\langle 0|v\rangle$ are the "Franck-Condon" factors. Then, the resonant electric field components $\mathcal{E}_{0,v}(t)\, e^{-i\omega_{0,v}t}$ have the amplitudes $\mathcal{E}_{0,v=1,\ldots,n}(t) = f_D(t)(C c_v^0)^*/\mu_{0,v}$ and $\mathcal{E}_{0,v=n+1,\ldots,n+m}(t) = f_P(t)(C' c_v^e)^*/\mu_{0,v}$, where for $0.1 < |c_v^{0(e)}| < 1$ the adiabaticity can be satisfied with the choice [12] $C = C' = 30/\tau = 1\ \text{ps}^{-1}$ and $f_{D[P]}(t) = \exp[-(t - t_{D(P)})^2/\tau^2]$, with $t_{D(P)} = t_{\text{end}}/2 \mp \tau$ and $t_{\text{end}} > 2\tau$. The pulses cause

the desired population transfer, since $\Omega_{0,v=1,\ldots,n}(0) = \mu_{0,v}\mathcal{E}_{0,v}(0) \propto (c_v^0)^* C f_D(0)$ and $\Omega_{0,v=n+1,\ldots,n+m}(t_{\rm end}) \propto (c_v^e)^* C f_P(t_{\rm end})$, as required by Eqs. (4)–(6). We can also compensate on the free evolution during the transfer, $\Omega_{0,v=n+1,\ldots,n+m}(t) \rightarrow \Omega_{0,v=n+1,\ldots,n+m}(t) e^{i\, t_{\rm end}\, \omega_{0,v}}$, so the final wave packet is obtained exactly at $t_{\rm end}$ [12].

In Fig. 2 we test Eq. (4), by considering a more general population transfer chain. This is composed of the transfer from state $|1\rangle$ to a linear combination of states $|4\rangle$ and $|5\rangle$, followed by the transfer from this superposition state to a superposition of the $|1\rangle$, $|2\rangle$, and $|3\rangle$ states, and back to the $|4\rangle$ plus $|5\rangle$ superposition. In the first transfer, we apply multimode Gaussian pulses for which the Rabi frequencies are given as $\Omega_{0,1}(t) = \mathcal{O}_{0,1} \exp[-(t - t_0)^2/\tau^2]$ and $\Omega_{0,4(5)}(t) = \mathcal{O}_{0,4(5)} \exp[-t^2/\tau^2]$, with $t_0 = 2\tau$ being the delay between the pulses and the coefficients of Eq. (6) are chosen to be $C = C' = 50/\tau$. The next transfers in the chain are realized by repeating the pulses with different central times and amplitudes $\mathcal{O}_{0,k}$, fitting Eq. (6).

We next investigate the INS states in Eq. (3), and form their *arbitrary* superposition, $|D^I\rangle = \sum_{k,k'} t_{kk'} |D^I_{kk'}\rangle$, that arrange $|D^I\rangle$ to correlate at $t = 0$ with some state $|\Psi\rangle = \sum_k c_k^0 |k\rangle$. From the definition in Eq. (3), we get that the vector of initial complex amplitudes $\mathbf{c}^0 \equiv (c_1^0, \ldots, c_n^0)$ is orthogonal $\mathbf{\Omega}^0 \cdot \mathbf{c}^0 = 0$ to the pump pulse $\mathbf{\Omega}^0 \equiv (\Omega_{0,1}, \ldots, \Omega_{0,n})$. Therefore, *none* of these INS become depopulated, which is to be contrasted with the MNS in Eq. (4), where $\mathbf{\Omega}^0 = (C\mathbf{c}^0)^* f_P$ in Eq. (6).

We now consider a population transfer from the state $|\Psi\rangle = c_1^0|1\rangle + c_2^0|2\rangle$ to the state $|3\rangle$, where $c_1^0 = \sqrt{0.7}$ and $c_2^0 = \sqrt{0.3}$. By using the time-independent amplitudes of the pump $\mathbf{\Omega}^0$ vector of the form

$$\begin{aligned} \mathcal{O}_{0,1}^0 &= (c_1^0)^* \cos\varphi + e^{i\,\varphi_{\rm mix}} c_2^0 \sin\varphi, \\ \mathcal{O}_{0,2}^0 &= (c_2^0)^* \cos\varphi - e^{i\,\varphi_{\rm mix}} c_1^0 \sin\varphi, \end{aligned} \tag{7}$$

we can switch from following the MNS of Eq. (4), obtained when $\varphi = 0$ or π, giving rise to a complete population transfer, to following the INS, obtained for $\varphi = \pi/2$, for which the transfer probability is zero. The "mixing" phase $\varphi_{\rm mix}$ can provide an additional control of this population transfer.

Figure 3 illustrates the φ-controlled transfer between these two limits. Regardless of the value of φ and $\varphi_{\rm mix}$, the intermediate state $|0\rangle$ is always empty. If $\varphi_{\rm mix} = \pi/2$ (solid lines), the final populations p_1, p_2, and p_3 of the states $|k = 1, 2, 3\rangle$ are flat in the vicinity of the INS ($\varphi = \pi/2$), and thus they are close to their initial values. If $\varphi_{\rm mix} = 0$ (thin lines), the populations p_1 and p_2 become, around $\varphi = \pi/2$, highly sensitive to small changes in the value of φ. This effect could be used to control the INS composition: by choosing $\Omega_{0,i} c_i^0 \approx -\sum_{k \neq i} \Omega_{0,k} c_k^0$, we can pick one state $|i\rangle$ as a *control element,* that adjusts the INS in the system.

In complete analogy with the INS, the FNS span a subspace defined by the vectors $|D^F\rangle = \sum_{l,l'} t_{ll'} |D^F_{ll'}\rangle$, where $t_{ll'}$ are *arbitrary* coefficients that make $|D^F\rangle$ to correlate at $t = t_{\rm end}$ with some state $|\Psi'\rangle = \sum_l c_l^e |l\rangle$. While the INS are never depopulated, the FNS *cannot* get populated by the dump pulses $\mathbf{\Omega}^e \equiv (\Omega_{0,n+1}, \ldots, \Omega_{0,n+m})$, because $\mathbf{\Omega}^e \cdot \mathbf{c}^e = 0$, where $\mathbf{c}^e \equiv (c_{n+1}^e, \ldots, c_{n+m}^e)$ results from a combination of the factors in Eq. (3) and the $t_{ll'}$ coefficients. This situation can again be contrasted with the MNS in Eq. (4), where $\mathbf{\Omega}^e = (C'\mathbf{c}^e)^* f_D$ in Eq. (6). During the

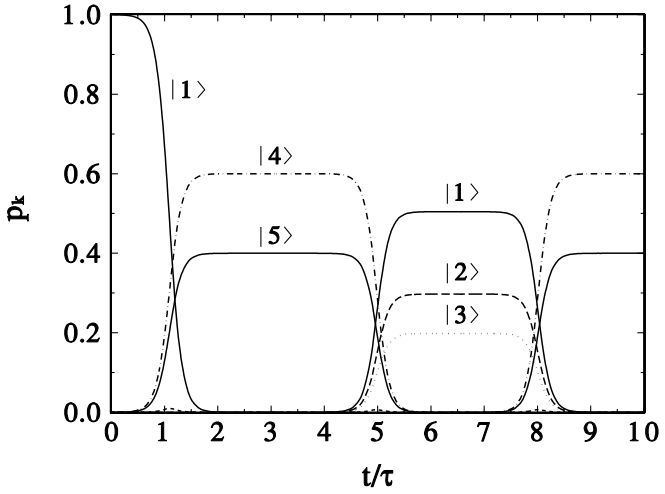

FIG. 2. Complete population transfers between various superpositions of the $|k\rangle$ $k = 1$–5 states. The lines are denoted by the symbols $|k\rangle$ for the respective populations $p_k = |c_k|^2$. All the phases of the c_k coefficients are chosen to be 0.

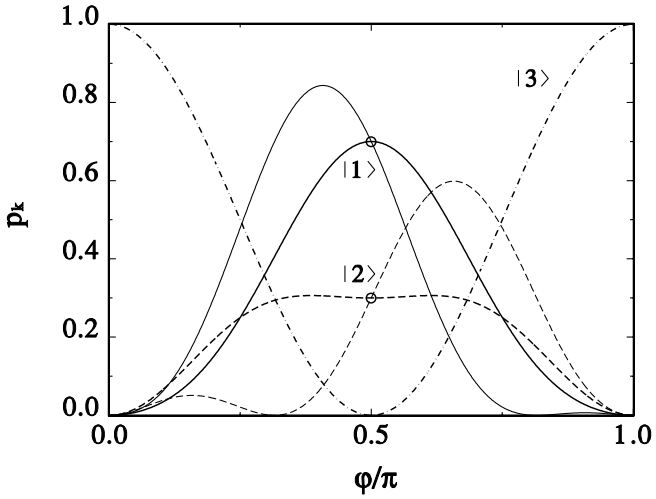

FIG. 3. Dependence of the final populations $p_k = |c_k|^2$ of the $|k\rangle$ states on the phase φ. The population transfer, from the state $\sqrt{0.7}|1\rangle + \sqrt{0.3}|2\rangle$ to the state $|3\rangle$, smoothly increases from 0% to 100% , as we change φ from $\pi/2$ to 0 or π, and go from the situation (INS) $\mathbf{\Omega}^0 \cdot \mathbf{c}^0 = 0$ to (MNS) $\mathbf{\Omega}^0 = (C\mathbf{c}^0)^* f_P$, respectively. The solid (thin) lines represent the situation with $\varphi_{\rm mix} = \pi/2$ ($\varphi_{\rm mix} = 0$).

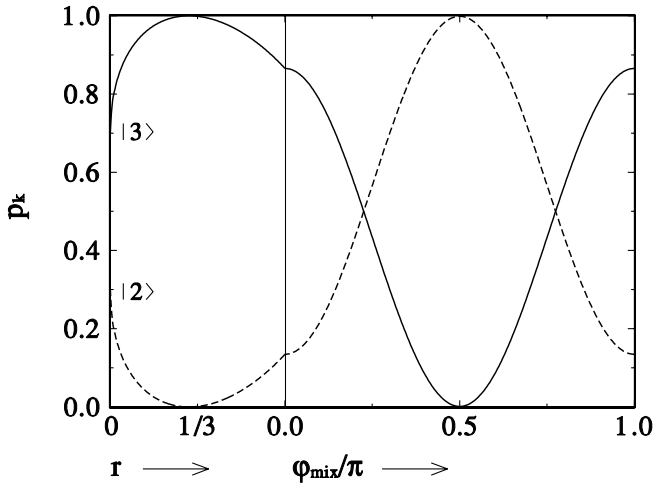

FIG. 4. Dependence of the final populations $p_k = |c_k|^2$ on the levels $|k\rangle$ ($k = 2, 3$) on the factor r (left side) and the phase $\varphi_{\rm mix}$ (right side). As we go from SAP ($r = 0$) to situations with nonzero *initial* population in states $|2, 3\rangle$ ($0 < r < 2/3$), the populations $p_{2,3}$ vary, but $p_2 + p_3 = 1$. Their ratio can be controlled by the mixing phase $\varphi_{\rm mix}$.

counterintuitive adiabatic evolution with the FNS, the initial state is fully depopulated and the intermediate state $|0\rangle$ never gets populated in all the cases, even when the FNS is initially partly populated. This implies that the final populations of the individual energy eigenstates $|k = n + 1, \ldots, n + m\rangle$, forming the FNS, are the result of a controlled *coherent mixing* between their initial and the transferred populations.

In Fig. 4, we present the final populations p_2 and p_3, obtained in transition from level $|1\rangle$ to a linear superposition of states $|D^F\rangle = c_2^e|2\rangle + c_3^e|3\rangle$, where $c_2^e = \sqrt{0.7}$, $c_3^e = \sqrt{0.3}$. The evolution starts from the initial state $|\Psi\rangle = \sqrt{1 - r}|1\rangle + \sqrt{r}|D^F\rangle$, combined from these states according to the parameter r. The chosen dump pulse $\mathbf{\Omega}^e$, with the Rabi amplitudes $\mathcal{O}_{0,2}^e = e^{i\varphi_{\rm mix}}c_3^e$, $\mathcal{O}_{0,3}^e = -e^{i\varphi_{\rm mix}}c_2^e$, is *always* orthogonal to the vector $\mathbf{c}^e$, so the $|D^F\rangle$ state is FNS. The mixing phase $\varphi_{\rm mix}$ determines, similarly as for the INS in Eq. (7), how the orthogonal vectors $\mathbf{c}^e$, $\mathbf{\Omega}^e$ are oriented one with respect to another. Thus it determines how the populations residing initially at the $|1\rangle$ and $|D^F\rangle$ states become mixed.

On the left hand side of Fig. 4, we keep $\varphi_{\rm mix} = 0$ and change r in the interval $0 < r < 2/3$, in order to show the transition from SAP [12], where $r = 0$ and $p_2/p_3 = |\Omega_{0,2}/\Omega_{0,3}|^2$, to some situations with FNS. We find that the population transfer from state $|1\rangle$ to the subspace spanned by states $|2\rangle$ and $|3\rangle$ is adiabatic and complete, and the ratio p_2/p_3 can be largely varied. On the right hand side of Fig. 4, we show that the mixing phase $\varphi_{\rm mix}$ can efficiently *control* the transfer of population to the individual states $|2\rangle$ or $|3\rangle$. This can dramatically change the ratio p_2/p_3, even for little populated FNS.

Additional control could be obtained, if the transitions of different components $\mathbf{c}^0$ are *split* in energy or time. Because of the unitary evolution, all the described processes are time reversible, so the system returns to the initial states if we reapply the used pulses in opposite order. This holds even if we study such a unitary evolution in systems with mixed states, described by density matrices, where new scenarios can be tested.

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