Dynamical Suppression of Unwanted Transitions in Multistate Quantum Systems

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Dynamical suppression of unwanted transitions in multistate quantum systems

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We propose a method to suppress unwanted transition channels and achieve perfect population transfer in multistate quantum systems by using composite pulse sequences. Unwanted transition paths may be present due to imperfect light polarization, misalignment of the quantization axis, spatial inhomogeneity of the trapping fields, off-resonant couplings, etc., or they may be merely unavoidable, e.g., due to perturbing excitations in molecules and solids. Compensation of separate or simultaneous deviations in polarization, pulse area, and detuning is demonstrated, even when these deviations are unknown, in three-state V and Ξ (ladder) systems and in a four-state Y system.

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Introduction. Experiments in many fields of quantum physics require well-defined quantum states and well-defined interactions. For example, the basic ingredient of the quantum computer is a well-defined qubit — a two-state quantum system. Real quantum systems, however, possess many states and special care is needed to isolate just two of them. In real and artificial atoms this is usually done with polarized laser light, carefully aligned with the quantization axis. However, unwanted transition channels may still be present, which reduce the fidelity of the operations. For example, when an ultracold atomic ensemble held in an optical dipole trap is addressed by right circularly polarized (σ^+) light, many atoms often "see" an admixture of σ^+ and σ^- light (i.e., elliptical polarization) since not all of them are exactly in the focus of the laser fields. Unwanted transitions may be present also due to imperfect polarization or alignment, off-resonant couplings, perturbing excitations in molecules and solids, etc.

In this Letter, we propose a simple and efficient technique for automatic compensation of such errors, even without knowing their magnitudes, which uses composite pulse sequences to dynamically suppress unwanted transitions, while simultaneously controlling the qubit in a robust way. We illustrate the technique in three- and four-state quantum systems forming linkages reminiscent of the letters V, Ξ and Y, as shown in Fig. 1; the technique is, however, applicable also to more complex linkage patterns. We demonstrate compensation of both independent and simultaneous variations in polarization, pulse area, and single-photon detuning (e.g., due to Stark shifts). The technique can also be used for compensation of unwanted two-photon and multi-photon detuning.

Composite pulse sequences have been used for several decades in nuclear magnetic resonance [1, 2], and since recently, in quantum information processing [3–6] and quantum optics [7–10] as a versatile control tool for quantum systems. Some of the basic ideas have been developed even earlier, in research on achromatic polarization retarders [11, 12]. While composite pulses have been used mainly for two-state quantum systems, there are also studies in three-state and multistate systems [10, 13].

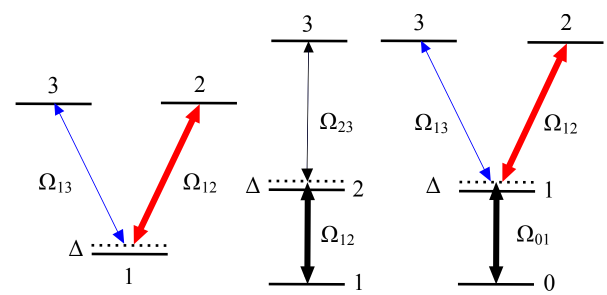

FIG. 1: (color online) V, Ξ(ladder) and Y systems.

A composite pulse is a sequence of pulses with well defined relative phases, which are used as control parameters in order to compensate imperfections in the excitation profile produced by a single pulse, or to shape the profile in a desired manner. The imperfections may be caused by an imprecise pulse area, undesirable frequency offset, or unwanted frequency chirp. Here we use the toolbox of composite pulses to design recipes for suppression of unwanted transition paths, which may turn a qubit or a simple three-state ladder system into a complex tree of states, with an unavoidable loss of efficiency.

V and Ξ systems. The dynamics of a coherently driven V system (Fig. 1, left), obeys the Schrödinger equation, $i\hbar\partial_t\mathbf{c}(t) = \mathbf{H}(t)\mathbf{c}(t)$, where the vector $\mathbf{c}(t) = [c_1(t), c_2(t), c_3(t)]^T$ contains the probability amplitudes of the three states. The Hamiltonian in the rotating-wave approximation (RWA) reads

$$\begin{aligned}\mathbf{H}_V(t) = (\hbar/2)\Delta(\Pi_{11} - \Pi_{22} - \Pi_{33}) + (\hbar/2)\\ \times \left[\Omega_{12}(t)e^{i\phi_{12}}\Pi_{12} + \Omega_{13}(t)e^{i\phi_{13}}\Pi_{13} + \text{h.c.}\right], \quad (1)\end{aligned}$$

where $\Delta = \omega_0 - \omega$ is the detuning between the laser carrier frequency ω and the Bohr transition frequency ω_0, and $\Pi_{jk} = |j\rangle\langle k|$. The magnitudes of the Rabi frequencies are $\Omega_{jk}(t) = |\mathbf{d}_{jk} \cdot \mathbf{E}(t)|/\hbar$, where $\mathbf{E}(t)$ is the envelope of the laser electric field and $\mathbf{d}_{jk}$ is the transition dipole moment of the respective transition $j \leftrightarrow k$; the phases of the Rabi frequencies are ϕ_{12} and ϕ_{13}. We assume that the two Rabi frequencies have the same time dependence $f(t)$ and we introduce the root-mean-square (RMS) peak Rabi frequency Ω and the mixing angle θ via $\Omega_{12}(t) = \Omega f(t)\cos\theta$ and $\Omega_{13}(t) = \Omega f(t)\sin\theta$. We assume hereafter that the composite sequence duration is shorter than the system's decoherence times.

An important example of a V system is the transition between the magnetic sublevel $m = 0$ (state $|1\rangle$) of a ground level with an angular momentum $j = 0$ or 1, and the magnetic sublevels $m = 1$ (state $|2\rangle$) and $m = -1$ (state $|3\rangle$) of an upper level with an angular momentum $j = 1$ driven by an elliptically polarized laser pulse. The latter is a superposition of two circularly polarized σ^+ and σ^- pulses [14]: then $\Omega_{12} = \Omega_+$ and $\Omega_{13} = \Omega_-$, the angle of rotation of the polarization ellipse is $\phi = (\phi_{12} - \phi_{13})/2$ and the ellipticity is $\varepsilon = \cos 2\theta$. The values $\theta = 0, \pi/4, \pi/2$ ($\varepsilon = 1, 0, -1$) correspond, respectively, to σ^+, linear and σ^- polarizations. The dynamics of the V system is similar to that of the Ξ system (Fig. 1, center), since the Hamiltonian of the latter is given by Eq. (1) by interchanging states $|1\rangle$ and $|2\rangle$.

Our objective is to transfer all population from state $|1\rangle$ to state $|2\rangle$ and completely suppress the excitation path $|1\rangle \leftrightarrow |3\rangle$ by using the phases ϕ_{1j} as control tool parameters. In the above example of magnetic sublevels, this can be achieved by a σ^+ polarized π pulse. However, if the polarization is not perfectly σ^+, then the atom will "see" some σ^- polarized light and the unwanted channel $|1\rangle \leftrightarrow |3\rangle$ will open up. We show below that composite pulses can compensate such an admixture of unwanted polarization, even without knowing its amount, and achieve perfect transfer $|1\rangle \to |2\rangle$.

The V system described by the Hamiltonian (1) can be transformed by the Morris-Shore transformation [15] into a decoupled state $|d\rangle = -e^{-i\phi_{13}}\sin\theta|2\rangle + e^{-i\phi_{12}}\cos\theta|3\rangle$ and a two-state system composed of state $|1\rangle$ and a coupled state $|c\rangle = e^{i\phi_{12}}\cos\theta|2\rangle + e^{i\phi_{13}}\sin\theta|3\rangle$ driven by the following Hamiltonian

$$\widetilde{\mathbf{H}}_2(t) = (\hbar/2)\{\Delta(\Pi_{11} - \Pi_{cc}) + [\Omega f(t)\Pi_{1c} + \text{h.c.}]\}. \quad (2)$$

We note that no population is trapped in the decoupled state permanently since its composition changes for each constituent pulse because the phases ϕ_{12} and ϕ_{13} change. The corresponding propagator can be expressed in terms of the complex Cayley-Klein parameters a and b (with $|a|^2 + |b|^2 = 1$) as [16]

$$\widetilde{\mathbf{U}} = \begin{bmatrix} a & b \\ -b^\star & a^\star \end{bmatrix}. \quad (3)$$

For resonant pulses ($\Delta = 0$), with RMS area $A = \int_{t_i}^{t_f} \Omega f(t)\mathrm{d}t$, the Cayley-Klein parameters are independent of the pulse shape: $a = \cos A/2$ and $b = -i\sin A/2$. For $\Delta \neq 0$, a and b depend on the pulse shape.

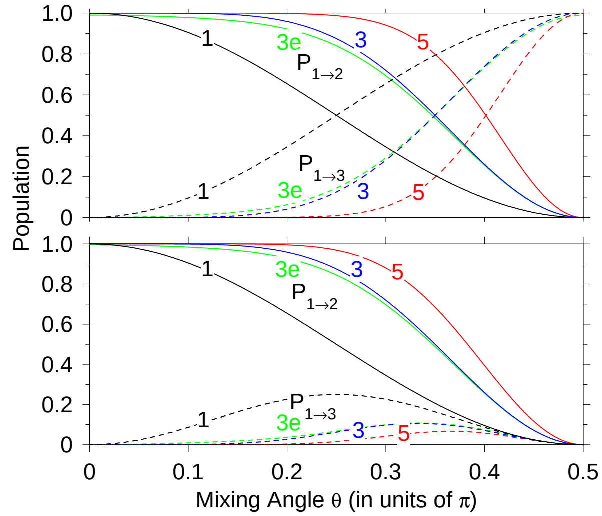

FIG. 2: (color online) Transition probabilities $P_{1\to 2}$ (solid) and $P_{1\to 3}$ (dashed) vs the mixing angle θ for a single resonant pulse with RMS area π and for composite sequences of N pulses (each with RMS area π). *Upper frame:* V system with phases $\boldsymbol{\phi}_{12} = (0, 2/3, 0)\pi$ and $\boldsymbol{\phi}_{13} = (0, 1, 1/3)\pi$ for 3 pulses, and $\boldsymbol{\phi}_{12} = (0, 1.411, 0.249, -0.432, -0.935)\pi$ and $\boldsymbol{\phi}_{13} = (0, 0.454, -0.632, 0.14, -0.514)\pi$ for 5 pulses. *Lower frame:* Ξ system with phases $\boldsymbol{\phi}_{12} = (0, 2/3, 1/6)\pi$ and $\boldsymbol{\phi}_{23} = (0, -2/3, 1/6)\pi$ for 3 pulses, and $\boldsymbol{\phi}_{12} = (0, -4, -1, 7, -4)\pi/10$ and $\boldsymbol{\phi}_{23} = (0, 4, 3, -5, 0)\pi/10$ for 5 pulses. The (3e) curves in both frames show the transition probabilities for 3 pulses when their phases experience random errors with a Gaussian distribution with a standard deviation of 0.05π.

The propagator in the original basis reads [10, 16, 17]

$$\mathbf{U}(\boldsymbol{\phi}) = \begin{bmatrix} a & b e^{i\phi_{12}} C & b e^{i\phi_{13}} S \\ -b^\star e^{-i\phi_{12}} C & a^\star C^2 + \zeta S^2 & (a^\star - \zeta)\, e^{-2i\phi} SC \\ -b^\star e^{-i\phi_{13}} S & (a^\star - \zeta)\, e^{2i\phi} SC & \zeta C^2 + a^\star S^2 \end{bmatrix}, \quad (4)$$

where $S = \sin\theta$, $C = \cos\theta$ and $\zeta = \exp[i\int_{t_i}^{t_f}\Delta(t)\mathrm{d}t/2]$. Complete population transfer $|1\rangle \to |2\rangle$ with a single pulse implies $|U_{21}| = 1$, i.e. $a = 0$, $|b| = 1$, $\theta = 0$. However, if $\theta \neq 0$, then the coupling between states $|1\rangle$ and $|3\rangle$ is nonzero and some population is unavoidably lost from state $|2\rangle$: transferred to $|3\rangle$ or left in $|1\rangle$.

Deviation of θ from 0 can be compensated to an arbitrary order by composite pulses. The propagator of a composite sequence of n pulses reads

$$\mathbf{U}^{(n)} = \mathbf{U}(\boldsymbol{\phi}_n)\cdots\mathbf{U}(\boldsymbol{\phi}_2)\mathbf{U}(\boldsymbol{\phi}_1), \quad (5)$$

where $\boldsymbol{\phi}_k = (\phi_{12}^{(k)}, \phi_{13}^{(k)})$ are the phase shifts of the k-th pulse in the sequence with $\mathbf{U}(\boldsymbol{\phi}_k)$ given by Eq. (4). The phases $\phi_{1j}^{(k)}$ are free parameters. We determine them by setting $P_{1\to 2} = |U_{21}^{(n)}|^2 = 1$ for $\theta = 0$ and nullifying the coefficients in the Taylor expansion of $P_{1\to 2}$ vs θ to the

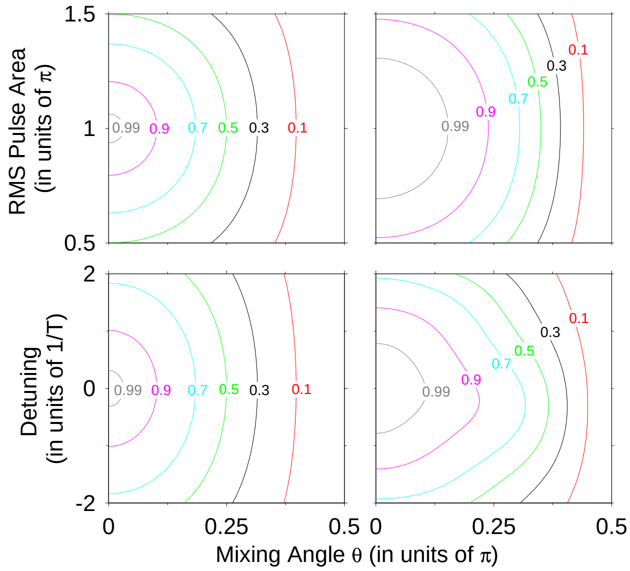

FIG. 3: (color online) Transition probability $P_{1\to2}$ in a V system. *Upper frames*: $P_{1\to2}$ vs the mixing angle θ and the RMS pulse area A for a single resonant pulse (upper left) and a composite sequence of three resonant pulses (upper right) with phases $\boldsymbol{\phi}_{12} = (0, 2/3, 0)\pi$ and $\boldsymbol{\phi}_{13} = (0, 1, 1/3)\pi$. *Lower frames*: $P_{1\to2}$ vs the mixing angle θ and the single-photon detuning Δ for a single rectangular pulse of duration T and RMS area $A = \pi$ (lower left) and a composite sequence of three rectangular pulses, each with duration T and RMS area π, and phases $\boldsymbol{\phi}_{12} = (0, 1/3, 0)\pi$ and $\boldsymbol{\phi}_{13} = (0, 2/3, 0)\pi$ (lower right).

highest possible order. Since the global phase is irrelevant, we take $\boldsymbol{\phi}_1 = \mathbf{0}$. This compensation is demonstrated in Fig. 2 (upper frame). For longer sequences (larger n), the transition profile $P_{1\to2}(\theta)$ broadens and the unwanted transition $|1\rangle \to |3\rangle$ is suppressed for a larger range of θ. Remarkably, for sufficiently long composite sequences, the transition $|1\rangle \to |3\rangle$ can be suppressed even if its coupling is larger than that of the transition $|1\rangle \to |2\rangle$, i.e. in the range $\theta > \pi/4$. Similarly, Fig. 2 (lower frame) shows suppression of the unwanted transition $|2\rangle \to |3\rangle$ in the Ξ system of Fig. 1 (center). The (3e) curves in Fig. 2 demonstrate the relative stability of results with respect to random errors in the phases.

Next, we have designed composite sequences which compensate *simultaneous* deviations in the mixing angle θ from 0 and the RMS pulse area A from π and (Fig. 3, top frames), and in the mixing angle θ from 0 and the single-photon detuning Δ from resonance (Fig. 3, bottom frames). In order to find the composite phases we use the Taylor expansion of $P_{1\to2}$ with respect to both θ and A (or Δ), and annul the coefficients of as many successive terms as possible, while requiring also $P_{1\to2} = 1$ for $\theta = 0$ and $A = \pi$ (or $\Delta = 0$). Similarly, simultaneous double compensation can also be done in the Ξ system. We note that even a triple compensation — vs θ, A and Δ — can be achieved in this manner; it is, however, more difficult to illustrate it.

Y system. The method for suppression of unwanted transitions is readily extended to more complex systems. Here we describe an extension of the V and Ξ systems to a Y-shaped system (Fig. 1, right). This system can arise from a three-state ladder due to control tools imperfections, like a two-state system is turned into a V or Ξ system. An example is found in coherent excitation of Rydberg states, e.g., in a cloud of ^{87}Rb atoms [18]. Additionally, the Y-system has the same coupling pattern as the well-known tripod system, in which three lower states are coupled to each other via two-photon transitions through a single upper state; this system is very important in applications using geometric phases [19].

The RWA Hamiltonian of the Y system reads

$$\mathbf{H}_Y(t) = \mathbf{H}_V(t) - (\hbar/2)\Delta\Pi_{00} + (\hbar/2)\left[\Omega_{01}(t)e^{i\phi_{01}}\Pi_{01} + \text{h.c.}\right], \tag{6}$$

where $\mathbf{H}_V(t)$ is the Hamiltonian (1) of the V system. The Rabi frequency $\Omega_{01}(t)$ of the additional transition $|0\rangle \leftrightarrow |1\rangle$, with phase ϕ_{01} (which provides an additional control parameter), should share the same time dependence $f(t)$ as the other two Rabi frequencies. In addition to the mixing angle θ in the V system, we introduce a second mixing angle ξ: $\Omega_{01}(t) = \Omega\sin\xi f(t)$, $\Omega_{12}(t) = \Omega\cos\xi\cos\theta f(t)$, and $\Omega_{13}(t) = \Omega\cos\xi\sin\theta f(t)$, where now $\Omega f(t) = \sqrt{\Omega_{01}(t)^2 + \Omega_{12}(t)^2 + \Omega_{13}(t)^2}$. Hereafter we take $\xi = \pi/4$, i.e., $\Omega_{01}(t)^2 = \Omega_{12}(t)^2 + \Omega_{13}(t)^2$. The couplings in the Y system in each interaction step can be caused by the simultaneous application of pairs of pulses from two lasers, one on the lower transition $|0\rangle \to |1\rangle$ and another (elliptically polarized) on the upper V system $|3\rangle \leftarrow |1\rangle \to |2\rangle$. The objective now is to transfer the population from state $|0\rangle$ to state $|2\rangle$ along the path $|0\rangle \to |1\rangle \to |2\rangle$, while suppressing the transition path $|1\rangle \to |3\rangle$. Mathematically, this requires $|U_{20}|^2 = 1$.

As in the V system, because the couplings share the same time dependence $f(t)$ and the Y system is on two-photon resonance, it can be transformed by the MS transformation into a set of two decoupled states and a two-state system. This allows us to obtain an exact analytic expression for the propagator in the original basis in terms of the Cayley-Klein parameters of the MS two-state system, similar to the one of Eq. (3) [10, 15–17]. The propagator $\mathbf{U}$ for the k-th pulse pair depends now on three phases: $\boldsymbol{\phi}_k = (\phi_{01}^{(k)}, \phi_{12}^{(k)}, \phi_{13}^{(k)})$.

Several conditions must be satisfied to achieve the desired transfer $|0\rangle \to |2\rangle$. When the mixing angle is $\theta = 0$ ($\Omega_{13} = 0$), this is achieved by a pair of simultaneous resonant pulses with RMS area $A = \int_{t_i}^{t_f}\Omega f(t)\mathrm{d}t = 2\pi$ [16]. As in the V system, unknown deviations in the interaction parameters can be compensated by a composite sequence of pulses. The propagator of a sequence of n pulse pairs is given by Eq. (5). Composite sequences are constructed in the same manner as for the V system: we expand

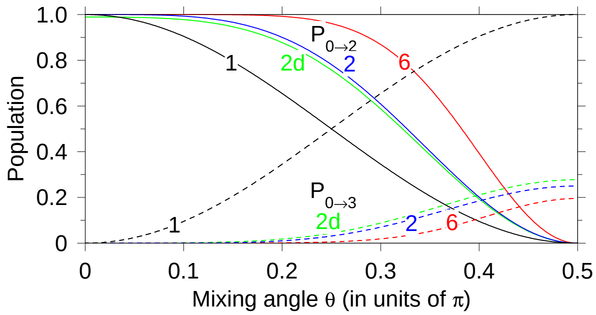

FIG. 4: (color online) Transition probabilities $P_{0\to2}$ (solid) and $P_{0\to3}$ (dashed) in a Y system vs the mixing angle θ for a single pulse pair with RMS area 2π, and for composite sequences of two and six pulse pairs (each with RMS area $A=\pi$), with phases: $\boldsymbol{\phi}_{01}=\boldsymbol{\phi}_{12}=(0,0)$ and $\boldsymbol{\phi}_{13}=(0,1)\pi$ for two pairs, and $\boldsymbol{\phi}_{01}=(0,0,-0.181,-0.181,-0.033,-0.033)\pi$, $\boldsymbol{\phi}_{12}=(0,0,-0.517,-0.517,-0.398,-0.398)\pi$, and $\boldsymbol{\phi}_{13}=(0,0.562,0.026,-1.554,0.393,0.238)\pi$ for six pairs. The (2d) curve shows the transition probabilities for the two pulse pairs when there is a random time delay of the $|0\rangle\to|1\rangle$ coupling, i.e. $\Omega_{01}(t)\to\Omega_{01}(t+\tau)$, where τ has a Gaussian distribution with a standard deviation of $0.1T$, where $A(T)=\pi$.

$P_{0\to2}=|U_{20}^{(n)}|^2$ in a Taylor series vs the relevant parameters and annul as many terms (in ascending order) as possible. Compensation vs the mixing angle θ is demonstrated in Fig. 4 for sequences of 2 and 6 pulse pairs; its relative stability to random time delays in the $|0\rangle\to|1\rangle$ coupling is also shown there. Simultaneous compensation of deviations in both the mixing angle θ and the RMS pulse area A is shown in Fig. 5 (upper frames), and in the mixing angle θ and the single-photon detuning Δ in Fig. 5 (lower frames) for sequences of 3 pulse pairs.

Discussion and Conclusion. Besides the proposed composite pulses technique, we note that other methods for suppressing unwanted transitions exist, e.g. dynamical decoupling for suppression of decoherence [20], which has been shown to be equivalent to the quantum Zeno effect [21] and extended to dynamically error-corrected gates (DCGs) [22, 23]. Our work differs from this approach in important details. "Bang-bang" decoupling keeps the system in a desired subspace by "strongly" coupling the qubit to the environment [21], i.e. by effectively projecting the total system onto the desired subspace. On the contrary, our approach relies on destructive interference of errors and it allows us to cancel unwanted couplings *of the same order of magnitude* as the desired ones, while DCGs can cancel perturbative errors.

We also note that examples of selective excitation by pulse trains have been demonstrated before [24–28]. The concept of our technique differs substantially from these because it allows for selective excitation to a desired state, and suppression of excitation to unwanted states when they are degenerate, i.e. we can *eliminate resonantly coupled states*, even *without knowing* how large the coupling to them is. The main reason for this advantage is that our technique relies on the differences in the phases of the unknown target and unwanted couplings.

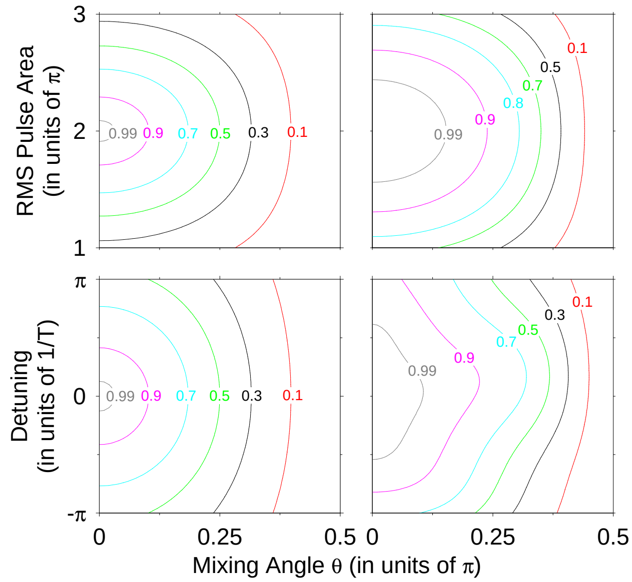

FIG. 5: (color online) Transition probability $P_{0\to2}$ for a Y system. *Upper frames*: $P_{0\to2}$ vs the mixing angle θ and the RMS pulse area A for a single resonant pulse pair (upper left) and a composite sequence of three resonant pulse pairs (upper right) with phases $\boldsymbol{\phi}_{01}=(0,1,-1/3)\pi$, $\boldsymbol{\phi}_{12}=(0,-2/3,1/3)\pi$, and $\boldsymbol{\phi}_{13}=(0,1,0)\pi$. *Lower frames*: $P_{0\to2}$ vs the mixing angle θ and the single-photon detuning Δ for a single pair of rectangular pulses with RMS area $A=2\pi$ (lower left) and a composite sequence of three pairs of rectangular pulses, each with RMS area 2π, and phases $\boldsymbol{\phi}_{01}=\boldsymbol{\phi}_{12}=(0,2/3,0)\pi$ and $\boldsymbol{\phi}_{13}=(0,1,0)\pi$.

The proposed technique is a simple and efficient method for robust population transfer and suppression of unwanted transition channels in multistate quantum systems. We have demonstrated it for three-state V and Ξ systems and four-state Y systems, but it can readily be adapted to more complex systems. Unwanted transition channels may be merely unavoidable (e.g. due to off-resonant couplings), or can be activated, for instance, by deviations in light polarization or geometric reasons. By suitably choosing the phases of the constituent pulses, the unwanted transitions can be suppressed with very high fidelity, while compensation of deviations in laser polarizations, intensities and detunings can be done simultaneously. The accuracy, the flexibility and the simplicity of the proposed technique make it a potentially important tool in applications requiring high control fidelity, such as quantum information processing and quantum optics.

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