Universality in exact quantum state population dynamics and control

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We consider an exact population transition, defined as the probability of finding a state at a final time that is exactly equal to the probability of another state at the initial time. We prove that, given a Hamiltonian, there always exists a complete set of orthogonal states that can be employed as time-zero states for which this exact population transition occurs. The result is general: It holds for arbitrary systems, arbitrary pairs of initial and final states, and for any time interval. The proposition is illustrated with several analytic models. In particular, we demonstrate that in some cases, by tuning the control parameters, a *complete* transition might occur, where a target state, vacant at $t = 0$, is fully populated at time τ.

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I. INTRODUCTION

The central goal of quantum control is the transfer of population from an initial state to a final target state [1,2]. Within the framework of coherent quantum control, focus has been primarily on designing specific laser-based scenarios that achieve this objective (for some bound-state examples, see, e.g., Refs. [3–5]), whereas within the framework of optimal control, focus has been on identifying control fields that reach this target, both computationally and experimentally.

Despite the enormous interest in this area, there are very few analytic control results about realistic systems. These include theorems such as that of Huang-Tarn-Clark [6], a theorem by Ramakrishna *et al.* on the dimensionality of the Lie algebra induced by the interaction between the system and the control field [7], and a theorem by Shapiro and Brumer [8], where control was shown to depend on the dimensionality of the controlled subspaces. As a consequence, any proven fundamental result adds considerably to the knowledge base (e.g., Ref. [9]). In this paper, we expose a universal feature of quantum *dynamics* that has significant implications for control. The focus here is on proving this dynamical result; future studies will direct control applications.

Specifically, consider an initial state $|\Psi(0)\rangle$ that evolves under a Hamiltonian H to yield the state $|\Psi(\tau)\rangle$ at time $t = \tau$. Of interest is the probability $P_I(0) = |\langle I|\Psi(0)\rangle|^2$ of the system being initially in state $|I\rangle$ undergoing a transition with probability $P_F(\tau) = |\langle F|\Psi(\tau)\rangle|^2$ to an orthogonal component $|F\rangle$ at time τ. We focus on the possibility of an exact quantum transition between these states defined as

$$P_F(\tau) = P_I(0) \tag{1}$$

(i.e., where the probability of observing state $|F\rangle$ at final time τ equals the probability of observing the state $|I\rangle$ initially).

In this paper, we prove that there always exists, for an arbitrary evolution operator and for an arbitrary time τ, a complete set $\{\Psi_k(0)\}$ of orthogonal states that undergo the exact state transition (1) from $|I\rangle$ to $|F\rangle$. For a given Hamiltonian, the magnitude of the associated $P_F(\tau)$ is determined by the choice of τ, $|I\rangle$, and $|F\rangle$. As examples, we obtain the set $\{\Psi_k(0)\}$ for some analytical models, and furthermore provide instances of *significant transfer*, defined as $P_F(0) \ll P_I(0)$.

While this universality might seem surprising, in what follows, we show that it arises directly from the unitarity of quantum evolution. Based on unitary evolution, the universal existence of exact quantum state transmission between different subspaces was demonstrated in Ref. [10], and cyclic quantum evolution in the theory of geometric phase was established in Ref. [11]. Unitarity is also at the heart of the no-cloning theorem, which is fundamental to quantum information science. Here, an inclusive theorem in quantum dynamics based on unitarity is derived, expected to be influential in quantum technologies. In particular, note that the dynamical principle is established here within the *same* Hilbert space, unlike Ref. [10], giving an approach that is propitious for a broad range of applications (e.g., for quantum computing [12]), coherent control of atomic and molecular processes [2], and laser control of chemical reactions in molecules [1].

II. UNIVERSALITY OF THE EXACT POPULATION TRANSITION

Consider an M-dimensional system (M can be infinite), spanned by the bases $\{|\alpha\rangle\}$ and described by density matrix ρ. The equality in Eq. (1) becomes

$$\mathrm{tr}\,[|F\rangle\langle F|\rho(\tau)] = \mathrm{tr}\,[|I\rangle\langle I|\rho(0)]. \tag{2}$$

We assume that the system is prepared in a pure state $\Psi(0)$ so that $\rho(0) = |\Psi(0)\rangle\langle\Psi(0)|$.

Proposition. There always exists a complete orthogonal set $\{\Psi_k(0)\}_\tau$, which depends on τ such that an exact population transition described by Eq. (1) or (2) takes place if the initially prepared state is a member of this set.

Proof. Assuming that at time $t = 0$ the state of the system is $\Psi(0)$, the left side of Eq. (2) can be written as

$$\begin{aligned}\mathrm{tr}\,[|F\rangle\langle F|\rho(\tau)] &= \mathrm{tr}\,[|F\rangle\langle F|U(\tau)\rho(0)U^\dagger(\tau)] \\ &= \langle\Psi(0)|U^\dagger(\tau)|F\rangle\langle F|U(\tau)|\Psi(0)\rangle \\ &= \langle\Psi(0)|U^\dagger(\tau)\mathcal{E}|I\rangle\langle I|\mathcal{E}U(\tau)|\Psi(0)\rangle \\ &= \mathrm{tr}\,[|I\rangle\langle I|\rho'(\tau)],\end{aligned} \tag{3}$$

where $U(\tau)$ is the time-evolution operator of the system, and we have introduced the exchange operator,

$$\mathcal{E} = |F\rangle\langle I| + |I\rangle\langle F| + \mathcal{E}_0, \tag{4}$$

satisfying $\mathcal{E}|I\rangle\langle I|\mathcal{E} = |F\rangle\langle F|$, with $\mathcal{E}_0 = \sum_{\alpha \neq I,F}^{M} |\alpha\rangle\langle\alpha|$. The exchange operator $\mathcal{E}$ swaps the states $|F\rangle$ and $|I\rangle$ while keeping other states intact. It is easy to prove that $\mathcal{E}^2 = 1$. We have also defined the auxiliary density matrix $\rho'(\tau) = |\Psi'(\tau)\rangle\langle\Psi'(\tau)|$, with $\Psi'(\tau) = W(\tau)\Psi(0)$ and $W(\tau) = \mathcal{E}U(\tau)$. This operator behaves similarly to the time-evolution operator. It is significant to note that the operator $W(\tau)$ is unitary, satisfying

$$W^\dagger(\tau)W(\tau) = U^\dagger(\tau)\mathcal{E}\mathcal{E}U(\tau) = 1. \tag{5}$$

As a unitary operator, $W(\tau)$ can be diagonalized to yield a complete set of orthonormal eigenvectors $\{\Psi_k(0)\}_\tau$ and exponential eigenvalues $\{\exp(i\phi_k)\}_\tau$. Any vector $\Psi_k(0)$ in the set thus obeys the eigenequation,

$$W(\tau)\Psi_k(0) = \exp(i\phi_k)\Psi_k(0). \tag{6}$$

Comparing Eqs. (2) with (3), we note that if the state of the system at time zero $\Psi(0)$ is one of the $\Psi_k(0)$'s in Eq. (6), then the equality $\mathrm{tr}[|I\rangle\langle I|\rho(0)] = \mathrm{tr}[|F\rangle\langle F|\rho(\tau)]$ in Eq. (2) holds. In other words, an exact population transition occurs between states $|I\rangle$ and $|F\rangle$ independent of the choice of these states other than that they are orthogonal. The result is also valid for the exchange operator $\mathcal{E} = e^{i\alpha}|F\rangle\langle I| + e^{i\beta}|I\rangle\langle F| + \mathcal{E}_0$, where α and β are real numbers. In this case, the unitary condition (5) translates to $W^\dagger(\tau)W(\tau) = U^\dagger(\tau)\mathcal{E}^\dagger\mathcal{E}U(\tau) = 1$, since $\mathcal{E}^\dagger$ may not be equal to $\mathcal{E}$.

The previous result is a fundamental attribute of quantum dynamics and should serve as a basic building block in quantum control theory (see also Refs. [10] and [11]). Given an arbitrary Hamiltonian at an arbitrary time τ and an arbitrary pair of states $|I\rangle$ and $|F\rangle$, $W(\tau)$ can be numerically diagonalized to obtain its eigenvalue spectrum and eigenstates. Employing these eigenstates as the time-zero system state leads to the exact transition $P_I(0) = P_F(\tau)$. This can be readily done for small systems, and in the following, we illustrate several eigenproblems where the spectrum of $W(\tau)$ can be analytically obtained. However, we emphasize that, unlike specific control scenarios, this result is *universal*, arising only from the fact that a unitary operator possesses a complete set of orthogonal eigenvectors. Indeed, even if the transition between $|I\rangle$ and $|F\rangle$ is not allowed, the theorem is still valid, although trivially producing $P_I(0) = P_F(0) = 1/2$.

Of particular interest in control scenarios are transitions, which we denote as *significant*, when $P_I(0) > P_F(0)$. Ideally, we seek exact transitions that are (what we term) *complete* [i.e., where $P_I(0) = 1$ and $P_F(0) = 0$ so that an initial state, fully populated at time zero, transfers its population to a target state at time τ]. Experience gained from individual sample cases that follow sheds light on the theorem and will allow one to assess future directions for control applications.

III. EXAMPLE: A TWO-LEVEL SYSTEM

We discuss three variants of the two-level-system (TLS) model. In the first *static* case, the theorem holds in a trivial way, though there is no actual population transition. In the second case, a weak time-dependent perturbation leads to a significant population transfer. The last example demonstrates that in a δ-kicked TLS a complete transition might take place. The unperturbed TLS model is described by the states $|0\rangle$ and $|1\rangle$ of energies E_0 and E_1, respectively. Taking into account different types of interactions, we explore next the transition between $|I\rangle = |0\rangle$ to $|F\rangle = |1\rangle$.

A. Static two-level system

Assuming for simplicity that $E_1 = 0$, we obtain the eigenstates of $W(\tau)$, $\Psi_\pm(0) = \frac{1}{\sqrt{2}}[|0\rangle \pm \exp(-iE_0\tau/2)|1]$ with eigenvalues $e^{i\phi_\pm}$, $\phi_+ = -E_0\tau/2$, and $\phi_- = -E_0\tau/2 + \pi$. This case is trivial, since there is no actual transition during the course of time. However, the dynamics is still Hamiltonian and hence Eq. (2) is valid.

B. Two-level system under a time-dependent perturbation

Consider next the two states $|0\rangle$ and $|1\rangle$ of energies E_0 and E_1, respectively, on-resonance with a periodic perturbation $H'(t) = \lambda V \cos\omega t$, where λ is a parameter characterizing the order of the perturbation expansion. Setting again $|I\rangle = |0\rangle$, $|F\rangle = |1\rangle$, we obtain the approximate eigenstates of $W(\tau)$ to first order of λ,

$$\Psi_\pm(0) \approx \frac{1}{\sqrt{2}\left(1 \pm \frac{r}{2}\cos\theta\right)}[-(r\cos\theta \pm 1)|0\rangle + |1\rangle], \qquad e^{i\phi_\pm} \approx \mp e^{\pm ir\sin\theta}, \tag{7}$$

where $re^{i\theta} = i\lambda\int_0^\tau ds e^{i(E_0-E_1)s}\cos(\omega s)\langle 1|V|0\rangle$; r and θ are real numbers. If $\cos\theta > 0$, we obtain the following inequality for the initial state $\Psi_+(0)$:

$$|\langle I|\Psi_+(0)\rangle| \approx \frac{1}{\sqrt{2}}\left(1 + \frac{r}{2}\cos\theta\right) > |\langle F|\Psi_+(0)\rangle| \approx \frac{1}{\sqrt{2}}\left(1 - \frac{r}{2}\cos\theta\right). \tag{8}$$

Since $|\langle I|\Psi_+(0)\rangle| > |\langle F|\Psi_+(0)\rangle|$, a significant transfer is realized here. For $\cos\theta < 0$, the same inequality holds for $\Psi_-(0)$. In both cases, the initial and final probabilities satisfy

$$P_I(0) = P_F(\tau) \approx \tfrac{1}{2}(1 + r|\cos\theta|). \tag{9}$$

Figure 1 demonstrates an exact population transfer in the present model, computed without approximation for the evolution of the TLS under a harmonic perturbation. Panel (a) demonstrates a significant transition, while for a different set of parameters, panel (b) shows that at specific times (or for a designed time-dependent field) a complete transition might take place, even for weak perturbations.

The analytic calculation Eqs. (7)–(9) exemplifies a significant transition [i.e., where $|\langle I|\Psi_k(0)\rangle| > |\langle F|\Psi_k(0)\rangle|$]. Having demonstrated that population transfer can be achieved, we further address the question, particularly relevant to control, of what is the maximum achievable significance, defined as $P_F(0) - P_I(0)$. For the TLS case, the significance equals $(-\mathrm{Tr}[\sigma_z\rho])$, which we want to maximize under the conditions that (a) the state is pure, and (b) an exact quantum transition is achieved at time τ. It can be shown that the condition for exact transition can be rewritten as $r_3 = -\vec{s}\cdot\vec{r}$,

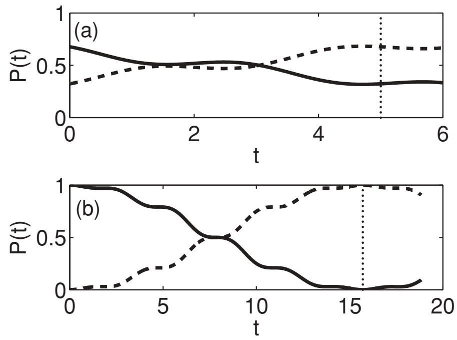

FIG. 1. The dynamics of a two-level system under a cosine perturbation. $E_1 - E_0 = \omega = 1,\ \lambda = 0.2,\ \langle 0|V|1\rangle = \langle 1|V|0\rangle = 1$. (a) An exact transition with $\tau = 5$. (b) A complete transition with $\tau = \pi/\lambda$; $P_I(t)$ (full line), $P_F(t)$ (dashed line). The dotted line marks the position of τ, where an exact transition takes place $P_I(0) = P_F(\tau)$.

where we define the vectors $\vec{s}(\tau) = \frac{1}{2}\mathrm{Tr}[U^\dagger(\tau)\sigma_z U(\tau)\vec{\sigma}]$ and $\vec{r} = \mathrm{Tr}[\rho\vec{\sigma}]$; $\sigma_{i=1,2,3}$ are the x,y,z Pauli matrices, respectively. If, for example, the component s_3 was equal to 1, the condition for an exact transition forces r_3 to be zero, and there is no initial state for which there is significant exact transfer. Assuming that $s_3 \neq 1$, on the other hand, the significance becomes maximal for the initial state,

$$\Psi(0) = \frac{1}{\sqrt{2(1-s_3)}}[(s_1 - is_2)|0\rangle + (1-s_3)|1\rangle]. \tag{10}$$

Note then, that generally, states that maximize the significance will *not* be eigenstates of $W(\tau)$. Thus, although in accord with the preceding theorem one achieves exact transitions, the ideal complete transition is obtained rarely. However, the proven theorem provides a new framework within which to modify the Hamiltonian to achieve transitions with increasingly larger significance.

C. Kicked two-level system

For the same TLS model, again with $|I\rangle = |0\rangle$ and $|F\rangle = |1\rangle$, consider a *nonperturbative* time-dependent Hamiltonian,

$$H = \begin{cases} \varpi(\sigma_z\cos\epsilon + \sigma_x\sin\epsilon), & 0 < t < \delta, \\ \omega\sigma_z, & t > \delta, \end{cases} \tag{11}$$

where σ_x, σ_y, and σ_z are the Pauli matrices. Here one can generically write the operator $W(\tau) = -i(\cos\theta + i\vec{n}\cdot\vec{\sigma}\sin\theta)$, where in this case the parameters are

$$\begin{aligned} \cos\theta &= \sin(\varpi\delta)\sin(\epsilon)\cos[\omega(\tau-\delta)], \\ n_z\sin\theta &= \sin(\varpi\delta)\sin(\epsilon)\sin[\omega(\tau-\delta)], \\ n_y\sin\theta &= -\cos(\varpi\delta)\sin[\omega(\tau-\delta)] \\ &\quad -\cos(\epsilon)\cos[\omega(\tau-\delta)]\sin(\varpi\delta), \\ n_x\sin\theta &= -\cos(\epsilon)\sin(\varpi\delta)\sin[\omega(\tau-\delta)] \\ &\quad +\cos(\varpi\delta)\cos[\omega(\tau-\delta)], \end{aligned} \tag{12}$$

with $n_x^2 + n_y^2 + n_z^2 = 1$. The two eigenstates of $W(\tau)$ are $\Psi_+(0) = \sqrt{\frac{1+n_z}{2}}(-i)e^{i\gamma}|0\rangle + \sqrt{\frac{1-n_z}{2}}|1\rangle$ with eigenvalue $-ie^{i\theta}$ and $\Psi_-(0) = \sqrt{\frac{1-n_z}{2}}ie^{i\gamma}|0\rangle + \sqrt{\frac{1+n_z}{2}}|1\rangle$ with eigenvalue $-ie^{-i\theta}$, where $\gamma = \arctan(n_x/n_y)$. Hence, adopting $\Psi_+(0)$ as the time-zero state, we obtain the results,

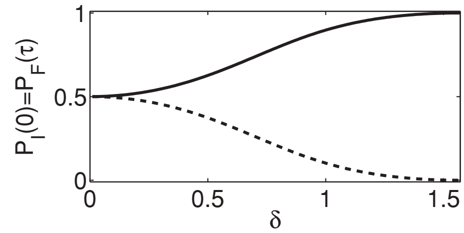

FIG. 2. The population $P_I(0) = P_F(\tau)$ for a kicked two-level model, showing that with proper tuning of parameters, a complete population transition can be achieved. $\tau = \pi$, $\epsilon = \pi/2$, $\omega = 1$, $\varpi = 1$. $|I\rangle = |0\rangle$ (full line); $|I\rangle = |1\rangle$ (dashed line).

$$P_F(\tau) = P_I(0) = \frac{1+n_z}{2}. \tag{13}$$

As an example, if $\epsilon = \pi/2$ and a strong pulse σ_x kicks at $t = 0$ such that $\varpi\delta \to \pi/2$, in the limit $\delta \to 0$, one gets that $n_z \to 1$, $\Psi_+(0) \to |0\rangle$, and $U(\tau)\Psi_+(0) \to |1\rangle$, which is a *complete* quantum transition from $|0\rangle$ to $|1\rangle$, satisfying $|\langle I|\Psi(0)\rangle| = 1$ and $|\langle F|\Psi(0)\rangle| = 0$. In Fig. 2, we show that by carefully tuning the interaction parameters (e.g., the delay time δ), one can achieve such a complete transition.

IV. COMPLETE TRANSITIONS: ADIABATIC EVOLUTION

We demonstrate here that a complete transition can take place in an adiabatically evolving system. Consider a time-dependent Hamiltonian $H(t)$. If it is varied sufficiently slowly, the evolution of the system is adiabatic, and the system occupies an (instantaneous) eigenstate of the Hamiltonian $H(t)$, provided the time-zero state $|\Psi(0)\rangle$ is an eigenstate of $H(0)$. If the state of the system at time τ $|\Psi(\tau)\rangle$ is orthogonal to the time-zero state, one can obtain a complete transition by setting $|I\rangle = |\Psi(0)\rangle$ and $|F\rangle = |\Psi(\tau)\rangle$, both eigenstates of $W(\tau)$. As an example, consider the magnetic Zeeman effect where a magnetic field splits the atomic (or molecular) degenerate levels, characterized by the magnetic quantum numbers M. The Hamiltonian is effectively given by

$$H = B(\epsilon)J_z + T(\epsilon)J_x, \tag{14}$$

where the second term $T(\epsilon)J_x$ is responsible for quantum transitions between different values of $M(= -J,\ldots,J)$. The time-dependent modulation is controlled by the parameter $\epsilon = \frac{\tau}{2} - t$, and we manipulate the magnetic field such that $T(\epsilon)$ $[B(\epsilon)]$ is an even [odd] function of ϵ, $T(\pm\frac{\tau}{2}) = 0$, and $B(0) > 0$. We now choose the initial state as $|I\rangle = |-J\rangle$, the lowest eigenstate of $H(0)$. If we control the evolution of $H(t)$ adiabatically from time 0 to τ, the state of the system at time τ becomes $|F\rangle = |J\rangle$, which is the lowest eigenstate of $H(\tau)$. Since $|J\rangle$ and $|-J\rangle$ are orthogonal, the quantum transition (2) is complete. The results of Ref. [13] may be an example of this scenario when $J = 1/2$.

V. SUPERPOSITION: EIGENSTATES IN A THREE-LEVEL SYSTEM

Finally, we address the following conceptual question: Can one achieve an exact population transition (1) with a superposition of the eigenstates of $W(\tau)$ as the time-zero state of the system? We explain next the conditions for this transfer by considering a three-level Hamiltonian $H = \Omega(|0\rangle\langle 1| + |1\rangle\langle 2| + \text{H.c.})$. If one chooses the initial and final states to be $|I\rangle = |0\rangle$ and $|F\rangle = |2\rangle$, the exchange operator $\mathcal{E} = |0\rangle\langle 2| + |2\rangle\langle 0| + |1\rangle\langle 1|$ commutes with H [14]. One can easily obtain the eigenstates and eigenvalues of $W(\tau) = \mathcal{E}U(\tau)$,

$$\begin{aligned}\Psi_a(0) &= -\frac{1}{\sqrt{2}}(|0\rangle - |2\rangle), \quad e^{i\phi_a} = -1,\\ \Psi_b(0) &= \frac{1}{2}(|0\rangle + \gamma_b|1\rangle + |2\rangle), \quad e^{i\phi_b} = e^{-i\sqrt{2}\Omega\tau},\\ \Psi_c(0) &= \frac{1}{2}(|0\rangle + \gamma_c|1\rangle + |2\rangle), \quad e^{i\phi_c} = e^{i\sqrt{2}\Omega\tau}.\end{aligned} \tag{15}$$

$\gamma_{b,c}$ are functions of τ and Ω, but their exact form is not important for the following discussion. Examining Eq. (15), it is obvious that there is no significant transition if the time-zero state is any of these three eigenstates, since $|\langle I|\Psi_k(0)\rangle| = |\langle F|\Psi_k(0)\rangle|$ for $k = a,b,c$. We show next that under some strict conditions, a superposition state can yield a complete transition. Since the set $\{\Psi_k(0)\}$ is a complete orthogonal set, a general time-zero state can be expanded as $\Psi(0) = \sum_{k=1}^{M} C_k\Psi_k(0)$, not necessarily an eigenstate of $W(\tau)$,

$$W(\tau)\Psi(0) = \sum_{k=1}^{M} C_k e^{i\phi_k}\Psi_k(0). \tag{16}$$

An exact transition can still take place at a specific time T, obeying the eigenvalue equation,

$$W(T)\Psi(0) = e^{i\phi(T)}\Psi(0). \tag{17}$$

This equality is satisfied if $C_k\{\exp(i\phi_k) - \exp[i\phi(T)]\} = 0$. Thus, for the contributing k coefficients $C_k \neq 0$, we obtain a set of conditions $\phi(T) = \phi_k + 2\pi K_k$, where K_k are arbitrary integers. This is a very restrictive condition when there are many nonzero coefficients. However, it may still be satisfied for particular systems with special symmetry. We exemplify this within the three-level model presented earlier [see Eq. (15)]. Assuming the following superposition state at time zero $\Psi(0) = C_a\Psi_a(0) + C_b\Psi_b(0)$, we obtain

$$W(\tau)\Psi(0) = -[C_a\Psi_a(0) - e^{-i\sqrt{2}\Omega\tau}C_b\Psi_b(0)], \tag{18}$$

which is generally not an eigenstate of $W(\tau)$. However, Eq. (17) is satisfied at the specific time $\tau = \pi/(\sqrt{2}\Omega)$ leading to

$$\begin{aligned}\langle I|\Psi(0)\rangle &= C_b/2 - C_a/\sqrt{2},\\ \langle F|\Psi(0)\rangle &= C_b/2 + C_a/\sqrt{2},\end{aligned} \tag{19}$$

manifesting a significant transition and a population transfer,

$$\begin{aligned}P_I(0) &= P_F(\pi/\sqrt{2}\Omega)\\ &= \frac{|C_a|^2}{2} + \frac{|C_b|^2}{4} - \frac{1}{2\sqrt{2}}(C_a^*C_b + C_b^*C_a).\end{aligned} \tag{20}$$

The transition can be made complete, depending on the superposition preparation coefficients C_b and C_a.

VI. CONCLUSIONS

We have proved the universality of exact quantum transitions, demonstrating that for a given Hamiltonian and a pair of states $|I\rangle$ and $|F\rangle$ in the associated Hilbert space, there always exists a complete set of orthogonal states that, when employed as the time-zero state of the system, leads to an exact population transition between the pair $|I\rangle$ and $|F\rangle$. This universal proposition is a fundamental feature of closed-system quantum dynamics, a promising building block in quantum control, and a result that should be readily demonstrable in sufficiently isolated systems, such as crossed molecular beams, cold atoms, and quantum dots that are weakly coupled to their environments. Applications to specific control studies are the subject of future work.

ACKNOWLEDGMENTS

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