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 being 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 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 \( \tau \).

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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 goal (for some bound state examples see, e.g., [3, 4, 5]), whereas within the framework of optimal control, focus has been on identifying control fields that achieve this goal, 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 Ramakrishnan et al. [7] 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., [9]). In this paper we expose a universal feature of quantum dynamics that has significant implications for control. The focus here is in the proving this dynamical result; future studies will be directed to 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 \( P_I(\tau) = \langle I | \Psi(0) \rangle^2 \) 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 \( \tau \). We focus on the possibility of an “exact quantum transition” between these states defined as

\[
P_F(\tau) = P_I(\tau),
\]

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

In this paper we prove that there always exists, for arbitrary evolution operator and for an arbitrary time \( \tau \), 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 \( \tau \), \(|I\rangle \), and \(|F\rangle \). As examples, we obtain the set \( \{\Psi_k(0)\} \) for some analytical models, and furthermore provide instances of significant transfer, defined by \( P_F(0) \ll P_I(0) \).

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

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 \( \rho \). The equality in Eq. (1) becomes

\[
\text{tr}[|F\rangle \langle F| \rho(\tau)] = \text{tr}[|I\rangle \langle I| \rho(0)].
\]

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 \( \tau \), such that an exact population transition described by Eqs. (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

\[
\text{tr}[|F\rangle \langle F| \rho(\tau)] = \text{tr}[|F\rangle \langle F| U(\tau) \rho(0) U^\dagger(\tau)]
\]

\[
= \langle \Psi(0) \rangle U^\dagger(\tau) |F\rangle \langle F| U(\tau) |\Psi(0)\rangle
\]

\[
= \langle \Psi(0) \rangle U^\dagger(\tau) E |I\rangle \langle I| E U(\tau) |\Psi(0)\rangle
\]
where $U(\tau)$ is the time evolution operator of the system, and we have introduced the exchange operator

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

satisfying $\mathcal{E} |I\rangle \langle I| \mathcal{E} = |F\rangle \langle F|$, with $\mathcal{E}_0 = \sum_{\alpha \neq I,F} \langle \alpha | \alpha \rangle$. 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 $\rho(k) = \exp(i\phi_k)\rho(k')\exp(-i\phi_k)$. Any vector $\Psi_k(0)$ in the set thus obeys the eigenequation

$$W(\tau)\Psi_k(0) = \exp(i\phi_k)\Psi_k(0).$$

Comparing Eq. (2) with Eq. (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 $\text{tr}(|I\rangle \langle I| \rho(\tau)) = \text{tr}(|F\rangle \langle F| \rho(\tau))$ 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 $\alpha$ and $\beta$ 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 above result is a fundamental attribute of quantum dynamics and should serve as a basic building block in quantum control theory (see also [10] and [12]). Given an arbitrary Hamiltonian at an arbitrary time $\tau$, and an arbitrary pair of states $|I\rangle$ and $|F\rangle$, $W(\tau)$ can be numerically diagonalized to obtain its eigenvalue spectrum and eigenstates, giving the states $|I\rangle$ and $|F\rangle$ between which the "exact transition" $P_I(0) = P_F(\tau)$ takes place. This can be readily done for small systems, and we illustrate below several simple eigen-problems 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. Of particular interest in control scenarios are transitions, which we denote as significant, when $P_I(0) > P_F(0)$. Ideally, in control scenarios, 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 $\tau$. Experience gained from individual sample cases below sheds light on the theorem and will allow one to assess future directions for control applications.

**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 delta-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 $|I\rangle = |0\rangle$ and $|F\rangle = |1\rangle$.

**I. Static two-level system.**— Assuming for simplicity that $E_0 = 0$, we obtain the eigenstates of $W(\tau)$, $\Psi_{\pm}(0) = \frac{1}{\sqrt{2}}(|0\rangle \pm \exp(-iE_0\tau/2) |1\rangle)$ 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 (2) is valid.

**II. 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 $\lambda$ 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 $\lambda$.

$$\Psi_{\pm}(0) \approx \frac{1}{\sqrt{2}}(1 \pm \frac{r}{2} \cos \theta) [-r \cos \theta \pm 1 |0\rangle + |1\rangle];$$

$$e^{i\phi} \approx \mp e^{\pm ir \sin \theta}$$

where $re^{i\theta} = i\lambda \int_0^\tau d\tau' \cos(\omega_0 - \omega_1)\varepsilon(\omega) \langle 1|V|0\rangle$; $r$ and $\theta$ are real numbers. If $\cos \theta > 0$ we obtain the following inequality for the initial state $\Psi_{+}(0)$

$$|\langle I|\Psi_{+}(0)\rangle| \approx \frac{1}{\sqrt{2}}(1 + \frac{r}{2} \cos \theta)$$

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

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

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

Figure [II] 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 (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 $P_F(\tau) - P_I(0)$. For the TLS case the significance equals...
Note then that generally, states that maximize the significance to modify the Hamiltonian to achieve transitions with incremental for the initial state for which there is significant exact transfer. Assuming generically write the operator (a) the state is pure, and (b) an exact quantum transition is achieved at time \( \tau \). It can be shown that the condition for an exact transition can be rewritten as 

\[
\sigma_z \sin \theta = -\cos(\epsilon) \sin(\omega(\tau - \delta)) \sin(\omega \delta),
\]

\[
n_x \sin \theta = -\cos(\epsilon) \sin(\omega \delta) \sin[\omega(\tau - \delta)],
\]

\[
+ \cos(\omega \delta) \cos[\omega(\tau - \delta)],
\]

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

\[
P_F(\tau) = P_F(0) = \frac{1 + n_x}{2}.
\]

As an example, if \( \epsilon = \pi/2 \) and a strong pulse \( \sigma_z \) kicks at \( t = 0 \) such that \( \omega \delta \to \pi/2 \), 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 \( \delta \), one can achieve such a complete transition. We next demonstrate that a complete transition can take place in general in an adiabatically evolving system.

**Complete transitions: Adiabatic evolution.**— 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 \( \tau \), \( |\Psi(\tau)\rangle \), 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

\[
H = B(\epsilon) J_z + T(\epsilon) J_x,
\]

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

Superposition: Eigenstates in a three-level system.— Finally, we address the following conceptual question: can one achieve an exact population transition 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 \( H = \Omega(|0 \rangle \langle 1| + |1 \rangle \langle 2| + h.c.) \). If one chooses the initial and final states to be \( |I \rangle = |0 \rangle \) and \( |F \rangle = |2 \rangle \), the exchange operator \( \mathcal{E} = \sum_{k} (|0 \rangle \langle 1| + |1 \rangle \langle 2| + h.c.) \) commutes with \( H \). One can easily obtain the eigenstates and eigenvalues of \( W(\tau) = \mathcal{E}U(\tau) \):

\[
\begin{align*}
\Psi_a(0) &= -\frac{1}{\sqrt{2}} (|0 \rangle - |2 \rangle); \quad e^{i\phi_a} = -1 \\
\Psi_b(0) &= \frac{1}{\sqrt{2}} (|0 \rangle + \gamma_b |1 \rangle + |2 \rangle); \quad e^{i\phi_b} = e^{-i\sqrt{2}\Omega \tau} \\
\Psi_c(0) &= \frac{1}{\sqrt{2}} (|0 \rangle + \gamma_c |1 \rangle + |2 \rangle); \quad e^{i\phi_c} = e^{i\sqrt{2}\Omega \tau}.
\end{align*}
\]

\( \gamma_{b,c} \) are functions of \( \tau \) and \( \Omega \) but their exact form is not important for the discussion below. Examining [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 \( \Psi(0) = \sum_k C_k \Psi_k(0) \), 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 be still satisfied for particular systems with special symmetry. We exemplify this within the three-level model presented above [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

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

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

\[
\begin{align*}
\langle I|\Psi(0)\rangle &= C_b/2 - C_a/\sqrt{2} \\
\langle F|\Psi(0)\rangle &= C_b/2 + C_a/\sqrt{2},
\end{align*}
\]

manifesting a significant transition and a population transfer

\[
P_f(t) = P_f(\pi/\sqrt{2}\Omega) = \frac{|C_a|^2}{2} + \frac{|C_b|^2}{4} - \frac{1}{2\sqrt{2}}(C_a^* C_b + C_b^* C_a). \tag{19}
\]

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

Conclusion.— 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, lead to an exact population transition between the pair \( |I \rangle \) and \( |F \rangle \). This universal proposition is a fundamental feature of quantum dynamics and a promising building block in quantum control. We have demonstrated the result analytically on the time-modulated two-level-system model, showing that in some cases a complete population transfer can be obtained. We have also shown that an adiabatic evolution can lead to a complete transition. Finally, we have analyzed exact population transitions in a superposition of states. Applications to specific control studies are the subject of future work.

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