Optimal Population Transfers in a Quantum System for Large Transfer Time

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***Abstract*—Transferringa quantum system to a final state with given populations is an important problem with applications to quantum chemistry and atomic physics. In this paper, we consider such transfers that minimize the L^2 norm of the control. This problem is challenging, both analytically and numerically. With the exception of the simplest cases, there is no general understanding of the nature of optimal controls and trajectories. We find that, by examining the limit of large transfer times, we can uncover such general properties. In particular, for transfer times large with respect to the time scale of the free dynamics of the quantum system, the optimal control is a sum of terms, each being a Bohr frequency sinusoid modulated by a slow amplitude, i.e., a profile that changes considerably only on the scale of the transfer time. Moreover, we show that the optimal trajectory follows a "mean" evolution modulated by the fast free dynamics of the system. The calculation of the "mean" optimal trajectory and the slow control profiles is done via an "averaged" two-point boundary value problem that we derive and which is much easier to solve than the one expressing the necessary conditions for optimality of the original optimal transfer problem.**

***Index Terms*—Optimal control, quantum systems.**

I. Introduction

STEERING a quantum system from its initial state to a given final state or a set of final states is one of the central problems in the control of such systems. While transfers to specific final states are very important for applications to quantum computing, for example, transfers to a set of final states with given populations are also important for many applications to quantum chemistry and atomic physics. Optimal control is a natural approach to such problems: Frequently, one desires to optimize some aspect of the transfer. For example, minimize the transfer time [1], maximize some measure of efficiency of the control in achieving its objective [2], or minimize some measure of the size of the control, for instance, its L^2 norm [3]–[5]. Moreover, the optimal control(s), singled out of all possible controls that achieve the objective, should have interesting properties tied to the structure of the given system.

In this paper, we consider only finite-dimensional quantum systems. In many applications, e.g., to molecular dynamics in quantum chemistry, these systems come from Galerkin (spectral) approximations of infinite-dimensional ones. Their dynamics are given by a finite dimensional Schrödinger equation

$$i\hbar\dot{\psi} = (H_0 + V\,u(t))\psi \tag{1}$$

where $\psi \in \mathbb{C}^N$ is the state and $\hbar$ is Planck's constant divided by 2π (which, from now on, will be set equal to 1). H_0 and V are Hermitian matrices, referred to as the system and control Hamiltonians, respectively. The control $u(t)$ is the amplitude of a laser (electromagnetic field). We consider a system with only one control, which is usually the case, but everything can be generalized to more controls. Let $\{E_i\}_{i=1,\ldots,N}$ be the eigenvalues of H_0 (energy levels of the quantum system) and $\{e_i\}_{i=1,\ldots,N}$ the orthonormal eigenvectors of H_0, referred to as energy eigenstates of the system, which furnish a basis for the state space. The quantities $\omega_{ij} \doteq E_i - E_j, i \neq j$, are the *Bohr frequencies* of the quantum system and the squared coefficients $|\psi_i|^2$ of the expansion $\psi = \sum_i \psi_i e_i$ are the *populations* of the energy eigenstates $e_i,\ \ i = 1, \ldots, N$.

We introduce the term *population transfer* to mean the transfer to a set of final states with *specified populations* $|\psi_i(T)|^2 = p_i,\ i = 1, \ldots, N$. In this paper, we consider *energy-optimal, exact* population transfers, that is we want to find a $u \in L^2([0,T])$, that minimizes the energy-like cost

$$\|u\|^2_{L^2([0,T])} = \int_0^T u^2(t)\,dt \tag{2}$$

and drives an initial state ψ_0 of system (1) to a final state $\psi(T)$ with *specified populations* $|\psi_i(T)|^2 = p_i$. We will refer to this as *optimal population transfer problem I* or **OPTP I**. This cost term for the control has been used extensively in the literature of optimal control of quantum systems as part of various objective functionals. It provides a measure of the energy spent to create the controlling field.

The maximum principle of optimal control provides necessary conditions for optimality in terms of the *Hamiltonian function*

$$H(\psi, \lambda, u) = \frac{1}{2}u^2 - i\lambda^*(H_0 + V\,u)\psi + i\psi^*(H_0 + V\,u)\lambda$$

where $\lambda \in \mathbb{C}^n$ is the costate vector. It is shown in the Appendix that solutions of the optimal control problem satisfy, $\lambda^*(t)\,\psi(t) = 0$. The optimal control u and the corresponding state and costate satisfy the equations

$$\dot{\psi} = \frac{\partial H}{\partial \lambda^*}, \quad \dot{\lambda} = -\frac{\partial H}{\partial \psi^*} \quad \text{and} \quad \frac{\partial H}{\partial u} = 0$$

which, given the expression for H, have the form

$$i\dot{\psi} = (H_0 + V\,u)\psi \tag{3}$$

$$i\dot{\lambda} = (H_0 + V\,u)\lambda \tag{4}$$

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$$u = i\,(\lambda^* V\psi - \psi^* V\lambda). \tag{5}$$

For convenience, we rewrite (3) and (4) using u from (5)

$$i\dot{\psi} = H_0\psi + i\,(\lambda^* V\psi - \psi^* V\lambda)\,V\psi \tag{6}$$

$$i\dot{\lambda} = H_0\lambda + i\,(\lambda^* V\psi - \psi^* V\lambda)\,V\lambda. \tag{7}$$

To these equations, one must append the boundary conditions

$$\psi(0) = \psi_0, \quad |\psi_i(T)|^2 = p_i, \quad \mathrm{Im}(\psi_i^*(T)\,\lambda_i(T)) = 0, \tag{8}$$

$\forall\, i = 1, \ldots, N$. The last of these equations are the transversality conditions at the endpoint. Their proof is also given in the Appendix. We will refer to the two-point boundary value problem comprised of (6)–(8) as *two-point boubdary value problem I* (**TPBVP I**) .

Relatively little work has been done on the problem of optimal *exact* population transfers. Analytically, it is a hard problem and explicit solutions are known only in few cases: For transfers between eigenstates in a 2-D system with two controls, the solution is given in [3] and [5] while [4] and [5] contain the solution for transfers between eigenstates of a 3-D system with four controls. In fact, both cases mentioned earlier are instances of a certain algebraic structure being present (the so-called $K + P$ structure) [4], [6]. It allows one to find analytic expressions for the control and the state in terms of the unknown initial costate, but the analytic determination of this unknown initial costate (and hence, the complete solution of the problem) is possible only for systems of small dimensionality because one needs to analytically compute matrix exponentials and solve transcendental equations. In dimensions higher than two, the presence of the $K + P$ structure requires unphysical numbers of controls and very special structure in the control Hamiltonians. For a two-state system with one control [3], it is possible to find an analytic expression for the control (in terms of unknown constants), but not for the state and so the problem has to be solved numerically from that point on. For higher-dimensional systems, no general properties of the optimal control and state trajectory are known.

Numerically, the TPBVP that expresses the necessary conditions of optimality becomes increasingly harder to solve as the dimension of the system or the transfer time grows. Dimension growth dramatically increases the computational cost of the numerical solution. More relevant to our paper is the issue of large transfer time. In many typical applications, the transfer time may be a few orders of magnitude larger than the time scale of the free evolution of the system. This may be necessary for the transfer to be possible or for the amplitude of the control to be within experimentally feasible limits. The problem here is the presence of two time scales in the solution, i.e., stiffness: There is the fast time scale of the free dynamics of the quantum system and the slow time scale of the transition. This creates the need for a very detailed numerical solution in order to guarantee good solution accuracy resulting in large computational times.

We found that the study of optimal population transfers for large transfer times offers some insight into the nature of optimal control and state trajectory as well as computational advantages in the numerical solution of the problem. The main conclusions of our paper are the following:

1) For generic population transfers and large-enough transfer times, the optimal control has the following, physically plausible form: It is a sum of sinusoids with frequencies equal to the Bohr frequencies ω_{ij} of the quantum system multiplied by slowly varying profiles, namely functions of $\frac{t}{T}$

$$u_{opt}(t) = \frac{i}{T}\mathrm{tr}\left(e^{iH_0 t}\,V\,e^{-iH_0 t}L\left(\frac{t}{T}\right)\right) + O\left(\frac{1}{T^2}\right)$$
$$= \frac{i}{T}\sum_{k\neq l} V_{kl}\,e^{i\omega_{kl}t}L_{lk}\left(\frac{t}{T}\right) + O\left(\frac{1}{T^2}\right)$$

where L is an anti-Hermitian matrix with zeros on the diagonal, whose entries are the profiles. This form is explicitly verified in all analytically solvable cases (where, because of the structure of the systems, there are no $O(\frac{1}{T^2})$ corrections) and it is, in fact, observed in numerical solutions.

2) Again, for generic population transfers and large enough transfer times, the optimal trajectory follows a slow "mean" evolution modulated by the fast free dynamics of the system. Quantitatively,

$$\psi(t) = e^{-iH_0 t}\,\bar{x}\left(\frac{t}{T}\right) + O\left(\frac{1}{T}\right)$$

where $\bar{x}$ denotes the "mean trajectory." The corrections are of higher order in an $\frac{1}{T}$ expansion. The slow mean evolution and the slow control profiles can be calculated by solving an "averaged" TPBVP (the term will be explained in Section II) in the fixed interval $[0, 1]$, irrespective of how large the transfer time T is.

Although the results above hold for large transfer times, one may use a solution of an optimal transfer problem obtained through our method for a large transfer time T as the first iterate in a continuation method solution of that optimal transfer problem for a smaller transfer time. The point is that the large transfer time limit both reveals the structure of the controls and serves as a good starting point for the solution of the problem. Indeed, the associated "averaged" TPBVP in $[0, 1]$ is much easier to solve numerically than the original problem.

Before we conclude this introductory section, we would like to address two points:

1) So far, we have only considered regular extrema of the OPTP I. Singular extrema [7] will not be considered in this paper. The reason is that the form of the singular extrema does not depend on the exact cost used in the minimization problem, and thus, the same singular extremum can be a local or even a global minimizer to many different cost functionals.
2) We briefly address the controllability question for system (1): It is controllable if, for every pair of initial and target states $(\psi_0,\ \psi_d)$, there exists a transfer time T and a measurable $u(t),\ t \in [0, T]$, such that the solution of (1) with $u(t)$ and $\psi(0) = \psi_0$ results into $\psi(T) = e^{i\varphi_d}\ \psi_d$, for some $\varphi_d \in S^1$ (in quantum mechanics, all states are defined modulo a total phase). Sufficiency conditions for controllability [8], [9] are based on the classical results [10]–[12].

In this paper, we would like to assume a somewhat strong form of controllability assumption as given, for example, in [9]. To state it, we need a few simple notions: A quantum system whose energy levels are all different from each other is called nondegenerate. Moreover, a system such that no two Bohr frequencies are the same is said to have no degenerate transitions. We also define the graph of an N-dimensional quantum system as a planar graph with N nodes, each representing an energy level, such that an edge connects the nodes i and j iff $V_{ij} \neq 0$. The positions of the graph nodes are not important, only the connectivity properties of the graph are (e.g., the graph is connected when there exists a sequence of edges connecting any two given nodes).

Controllability Assumption: The system (1) is nondegenerate, has no degenerate transitions, and the graph of V is connected.

Then [9], (1) is controllable. As a matter of fact, this is the generic situation for controllability of (1).

The rest of this paper is organized as follows: In Section II, we consider optimal population transfers for an "averaged" control system that approximates (1) to first order in a $\frac{1}{T}$ perturbation expansion. Section III contains our main results, namely that for large transfer times, the solution to the original optimal control problem is approximated by the solution of the optimal population transfer problem for the averaged system. This implies that the optimal control for the original problem belongs to the class of controls used to transform the original system to the averaged one and this provides a useful characterization of it. We demonstrate our approach with some examples in Section IV. The proof of our result is contained in Section V. Section VI concludes.

II. Optimal Population Transfers for an Averaged System

In this section, we introduce a special form for the control in (1), a sum of sinusoids with frequencies equal to the Bohr frequencies of the quantum system multiplied by slowly varying profiles, that is functions of $\frac{t}{T}$. We then proceed to "average out" the dynamics in the time scale of the free evolution of the system (this time scale is set by the Bohr frequencies), which is fast compared to the transfer time T. This leaves us with an "averaged" control system whose evolution approximates that of the original under the special form of the control introduced. The motivation for this lies in the following: We set up a corresponding OPTP for the averaged system, whose cost approximates the cost (2). We will show in Section V how solutions to this OPTP approximate solutions to our original OPTP I, to first order in an $O(\frac{1}{T})$ expansion, proving the results described in the introduction. We begin with the change of variable

$$x = e^{iH_0 t}\psi \tag{9}$$

in (1). In physics, this is referred to as transforming to the *interaction picture* or the *rotating frame*. The time evolution of the new variable x is due entirely to the control, because the free evolution has been accounted for. In terms of the new variable, (1) becomes

$$i\dot{x} = u\,F(t)x \tag{10}$$

where

$$F(t) \doteq e^{iH_0 t}\,V\,e^{-iH_0 t}. \tag{11}$$

Note the appearance of the Bohr frequencies in the matrix elements of F

$$F_{ij}(t) = V_{ij}e^{i(E_i - E_j)t} = V_{ij}e^{i\omega_{ij}t}.$$

We adopt the following form for the control $u(t)$

$$u(t) = \varepsilon\left(u_0(\varepsilon t) + \sum_{i\neq j}^{N} e^{i\omega_{ij}t}\,u_{ji}(\varepsilon t)\right) \tag{12}$$

where $u_{ji}^* = u_{ij}$ and u_0 is real, so that u is real. u_{ji} is a complex "envelope" that multiplies a sinusoid with frequency equal to the Bohr frequency for the transition i to j. The value of ε will be given shortly. We introduce $u(t)$ from (12) in (10) and rewrite (10) in component form

$$i\dot{x}_i = \varepsilon\left(u_0(\varepsilon t) + \sum_{k\neq l} e^{i\omega_{kl}t}\,u_{lk}(\varepsilon t)\right)\sum_j e^{i\omega_{ij}t}V_{ij}x_j. \tag{13}$$

We approximate (13) for small ε using averaging (in quantum mechanics, averaging is often used under the pseudonym *rotating wave approximation*). In averaging, one considers equations of the form

$$\dot{w} = \varepsilon f(w, t, \varepsilon) \tag{14}$$

such that the limit

$$f_{av}(w) \doteq \lim_{\tau\to\infty}\frac{1}{\tau}\int_t^{t+\tau} f(w, t', 0)\,dt'$$

exists and is independent of t. f must also be a C^2 function of its arguments with bounded derivatives up to second order. A standard averaging theorem [13, Ch. 8] guarantees that, for sufficiently small ε, the solution $\bar{w}(t)$ of

$$\dot{\bar{w}} = \varepsilon f_{av}(\bar{w})$$

with $\bar{w}(0) - w(0) = O(\varepsilon)$ satisfies $\bar{w}(t) - w(t) = O(\varepsilon)$ for $t \sim O(\frac{1}{\varepsilon})$. We need a similar result for equations of the form

$$\dot{w} = \varepsilon g(w, t, \varepsilon t, \varepsilon) \tag{15}$$

with only the $O(1)$ time-scale dynamics averaged. We apply the method of averaging to this equation, as follows: Define $w_0 = \varepsilon t$ and substitute w_0 for εt in (15). Then, consider the

system

$$\dot{w} = \varepsilon g(w, t, w_0, \varepsilon)$$
$$\dot{w_0} = \varepsilon$$

and apply averaging to it. The resulting averaged form of (15) is now

$$\dot{\bar{w}} = \varepsilon g_{av}(\bar{w}, \varepsilon t)$$

where

$$g_{av}(w, w_0) \doteq \lim_{\tau \to \infty} \frac{1}{\tau} \int_t^{t+\tau} g(w, t', w_0, 0)\, dt'.$$

We now apply this to (13). Since the time average of $e^{i\omega t}$ is zero for $\omega \neq 0$ and 1 for $\omega = 0$, only terms with no exponentials will contribute to the averaged equation. Letting $\bar{x}$ be the averaged x, the averaged form of (13) is

$$i\dot{\bar{x}}_i = \varepsilon \left(V_{ii}\, u_0(\varepsilon t)\, \bar{x}_i + \sum_{j \neq i} V_{ij}\, u_{ij}(\varepsilon t)\, \bar{x}_j \right). \tag{16}$$

Letting $\varepsilon = \frac{1}{T}$ and rescaling time to $s = \varepsilon t = \frac{t}{T}$, (16) becomes

$$i\frac{d\bar{x}_i}{ds} = V_{ii}\, u_0(s)\, \bar{x}_i + \sum_{j \neq i} V_{ij}\, u_{ij}(s)\, \bar{x}_j \tag{17}$$

or, in vector form

$$i\frac{d\bar{x}}{ds} = \tilde{V}[u_0, u_{ij}]\, \bar{x} = \begin{pmatrix} V_{11}u_0 & V_{12}u_{12} & \cdots \\ V_{21}u_{21} & V_{22}u_0 & \cdots \\ \vdots & \vdots & \ddots \end{pmatrix} \bar{x}.$$

By construction, every solution to (17) with controls $u_0(s)$ and $u_{ij}(s)$ and initial state $\bar{x}(0) = \psi(0)$, provides a solution to (1) with $u(t)$ given by (12) and initial condition $\psi(0)$, correct up to $O(\varepsilon)$ terms for a time interval of size $\frac{1}{\varepsilon} = T$. It is shown in the Appendix that (17) is controllable on account of the controllability assumption on the original system.

Since our goal is to relate optimal transfers of the original system to optimal transfers of the averaged one, we must find an objective for the averaged system that approximates $\|u\|^2_{L^2([0,T])}$. So, we compute $\|u\|^2_{L^2([0,T])}$ for $u(t)$ given by (12), with $\varepsilon = \frac{1}{T}$

$$\begin{aligned} &\int_0^T u^2(t)\, dt \\ &= \frac{1}{T^2} \int_0^T \left(u_0\left(\frac{t}{T}\right) + \sum_{i \neq j} e^{i\omega_{ij}t}\, u_{ji}\left(\frac{t}{T}\right) \right)^2 dt \\ &= \frac{1}{T} \int_0^1 \left\{ u_0^2(s) + 2\sum_{i \neq j} e^{i\omega_{ij}Ts}\, u_{ji}(s)\, u_0(s) \right. \\ &\quad \left. + \sum_{i \neq j} \sum_{k \neq l} e^{i(\omega_{ij}+\omega_{kl})Ts}\, u_{ji}(s)\, u_{lk}(s) \right\} ds \\ &= \frac{1}{T} \int_0^1 \left[\sum_{i \neq j} u_{ij}(s)\, u_{ji}(s) + u_0^2(s) \right] ds + O\left(\frac{1}{T^2}\right). \end{aligned}$$

The last line is the result of separating the integrals into two kinds, those without exponentials, which are explicitly retained, and those with, which can easily be seen to scale like $O(\frac{1}{T})$ after a partial integration. Indeed, for any differentiable f,

$$\begin{aligned} \int_0^1 e^{i\omega Ts}\, f(s)\, ds &= \frac{1}{i\omega T} \int_0^1 (e^{i\omega Ts})'\, f(s)\, ds \\ &= \frac{1}{i\omega T} \left\{ e^{i\omega T}\, f(1) - f(0) - \int_0^1 e^{i\omega Ts}\, f'(s)\, ds \right\}. \end{aligned}$$

We pose the following optimal control problem for the averaged system: Find controls $u_0(s)$, $u_{ij}(s)$, $s \in [0,1]$, that minimize

$$\begin{aligned} &\int_0^1 \left[\sum_{i \neq j} u_{ij}(s)\, u_{ji}(s) + u_0^2(s) \right] ds \\ &\quad = \int_0^1 \left[\sum_{i \neq j} |u_{ij}(s)|^2 + u_0^2(s) \right] ds \end{aligned} \tag{18}$$

and drive an initial state $\bar{x}(0) = \psi_0$ of system (17) to a target population distribution $|\bar{x}_i(1)|^2 = p_i,\ i = 1, \ldots, N$. We will refer to this as **OPTP II**.

The necessary conditions for optimality are derived from the Hamiltonian function

$$\begin{aligned} &\bar{H}(\bar{x}_i, \bar{z}_i, u_0, u_{ij}) \\ &\quad = \frac{1}{2}u_0^2 + \frac{1}{2}\sum_{i \neq j} |u_{ij}|^2 - i\bar{z}^* \tilde{V}[u_0, u_{ij}]\, \bar{x} + i\bar{x}^* \tilde{V}[u_0, u_{ij}]\, \bar{z} \end{aligned}$$

and have the form

$$i\frac{d\bar{x}_i}{ds} = V_{ii}\, u_0\, \bar{x}_i + \sum_{j \neq i} V_{ij} u_{ij}\, \bar{x}_j \tag{19}$$

$$i\frac{d\bar{z}_i}{ds} = V_{ii}\, u_0\, \bar{z}_i + \sum_{j \neq i} V_{ij} u_{ij}\, \bar{z}_j \tag{20}$$

$$u_{ij} = iV_{ji}\, (x_i z_j^* - z_i x_j^*)$$

$$u_0 = i\sum_i V_{ii}(\bar{x}_i \bar{z}_i^* - \bar{z}_i \bar{x}_i^*). \tag{21}$$

We rewrite (19) and (20) using u_0 and u_{ij} from (21)

$$\begin{aligned} \frac{d\bar{x}_i}{ds} &= \sum_{j \neq i} |V_{ij}|^2 (\bar{x}_i \bar{z}_j^* - \bar{z}_i \bar{x}_j^*)\, \bar{x}_j \\ &\quad + \sum_k V_{kk} (\bar{x}_k \bar{z}_k^* - \bar{z}_k \bar{x}_k^*)\, V_{ii} \bar{x}_i \end{aligned} \tag{22}$$

$$\begin{aligned} \frac{d\bar{z}_i}{ds} &= \sum_{j \neq i} |V_{ij}|^2 (\bar{x}_i \bar{z}_j^* - \bar{z}_i \bar{x}_j^*)\, \bar{z}_j \\ &\quad + \sum_k V_{kk} (\bar{x}_k \bar{z}_k^* - \bar{z}_k \bar{x}_k^*)\, V_{ii} \bar{z}_i. \end{aligned} \tag{23}$$

The corresponding boundary conditions are given by

$$\bar{x}(0) = \psi_0, \quad |\bar{x}_i(1)|^2 = p_i, \quad \mathrm{Im}(\bar{x}_i^*(1)\,\bar{z}_i(1)) = 0. \tag{24}$$

The proof of the transversality conditions at the endpoint $s = 1$, is done exactly the same way as for OPTP I in the Appendix. Similarly to the state and costate of OPTP I, it can be shown that $\bar{z}^*(s)\bar{x}(s) = 0\ \forall s \in [0,1]$. We will refer to the TPBVP comprised of (21) and (22) along with (24) as **TPBVP II**. From (21), we see that all u_{ij} with $V_{ij} = 0$ are identically zero, as they should. We can also show that $u_0 = 0$ and simplify the right sides of (22) and (23). To this end, we define the anti-Hermitian matrices $L \doteq \bar{x}\bar{z}^* - \bar{z}\bar{x}^*$, and $K = K(L)$ given by

$$K_{ij} \doteq |V_{ij}|^2 L_{ij}, \quad i \neq j \tag{25}$$

$$K_{ii} \doteq V_{ii} \sum_k V_{kk} L_{kk}. \tag{26}$$

With these definitions, (22) and (23) read simply as

$$\frac{d\bar{x}}{ds} = K(L)\bar{x} \tag{27}$$

$$\frac{d\bar{z}}{ds} = K(L)\bar{z}. \tag{28}$$

It is easy to see that L satisfies the differential equation

$$\frac{dL}{ds} = [K(L), L]. \tag{29}$$

The iith component of this equation reads

$$\begin{aligned}\frac{dL_{ii}}{ds} &= \sum_j K_{ij}L_{ji} - L_{ij}K_{ji} \\ &= K_{ii}L_{ii} + \sum_{j\neq i} K_{ij}L_{ji} - L_{ii}K_{ii} - \sum_{j\neq i} L_{ij}K_{ji} \\ &= \sum_{j\neq i} |V_{ij}|^2 (L_{ij}L_{ji} - L_{ij}L_{ji}) = 0\end{aligned}$$

and so

$$L_{ii}(s) = L_{ii}(1) = -2i\,\mathrm{Im}(\bar{x}_i^*(1)\,\bar{z}_i(1)) = 0. \tag{30}$$

This shows that $K_{ii}(s) = L_{ii}(s) = 0$ and $u_0(s) = 0$, and so (22) and (23) simplify to

$$\frac{d\bar{x}_i}{ds} = \sum_{j\neq i} |V_{ij}|^2 (\bar{x}_i\bar{z}_j^* - \bar{z}_i\bar{x}_j^*)\,\bar{x}_j \tag{31}$$

$$\frac{d\bar{z}_i}{ds} = \sum_{j\neq i} |V_{ij}|^2 (\bar{x}_i\bar{z}_j^* - \bar{z}_i\bar{x}_j^*)\,\bar{z}_j. \tag{32}$$

We will refer to this set of equations [along with (24)] as TPBVP II as well.

III. Main Results

In this section, we make the connection between solutions of the TPBVPs I and II in the large-T limit.

Theorem 1: Let $(\bar{x}(s),\ \bar{z}(s))$ be a solution of TPBVP II over $[0,1]$. Define $\psi(t),\ \lambda(t),$ and $u(t),\ t \in [0,T]$, by

$$\psi(t) = e^{-iH_0 t}\,\bar{x}\left(\frac{t}{T}\right) \tag{33}$$

$$\lambda(t) = \frac{1}{T}\,e^{-iH_0 t}\,\bar{z}\left(\frac{t}{T}\right) \tag{34}$$

$$\begin{aligned}u(t) &= \frac{i}{T}\,\mathrm{tr}\left(e^{iH_0 t}\,V\,e^{-iH_0 t}L\left(\frac{t}{T}\right)\right) \\ &= \frac{i}{T}\sum_{kl} V_{kl}\,e^{i\omega_{kl}t}L_{lk}\left(\frac{t}{T}\right).\end{aligned} \tag{35}$$

Then, for large-enough T, $\psi(t)$ satisfies the necessary conditions of optimality (3)–(8), up to terms of order $O(\frac{1}{T})$ and $\lambda(t)$ and $u(t)$ up to terms of order $O(\frac{1}{T^2})$.

Thus, solutions of the TPBVP II provide *approximate* solutions to the TPBVP I for large transfer times T. A natural question to ask then is whether these approximate solutions to TPBVP I are, in fact, *approximations to solutions* of I, in the large-T limit. This is answered positively by the following theorem.

Theorem 2: Let ψ_0 and $\{p_i\}_{i=1,\ldots,N}$ be an initial state and a target population of system (1), respectively, and let $(\bar{x}(s),\ \bar{z}(s))$ be a solution of TPBVP II over $[0,1]$. Then, for almost all pairs $(\psi_0, \{p_i\})$, a solution of TPBVP I exists, for large enough T, of the form

$$\psi(t) = e^{-iH_0 t}\,\bar{x}\left(\frac{t}{T}\right) + O\left(\frac{1}{T}\right) \tag{36}$$

$$\lambda(t) = \frac{1}{T}e^{-iH_0 t}\,\bar{z}\left(\frac{t}{T}\right) + O\left(\frac{1}{T^2}\right). \tag{37}$$

The corresponding control has the form

$$\begin{aligned}u(t) &= \frac{i}{T}\mathrm{tr}\left(e^{iH_0 t}\,V\,e^{-iH_0 t}L\left(\frac{t}{T}\right)\right) + O\left(\frac{1}{T^2}\right) \\ &= \frac{i}{T}\sum_{kl} V_{kl}\,e^{i\omega_{kl}t}L_{lk}\left(\frac{t}{T}\right) + O\left(\frac{1}{T^2}\right).\end{aligned} \tag{38}$$

The set of pairs $(\psi_0, \{p_i\})$ of initial states and target populations for which a solution of TPBVP II provides a solution of I according to (36) and (37) is open and full measure in the corresponding product manifold of initial states $\times$ final populations.

According to this theorem, solutions to the TPBVP II approximate solutions to I, for large transfer times, in the sense of (36)–(38), for almost every population transfer. In other words, all the local minima of OPTP II approximate local minima for the OPTP I according to (36)–(38). The question arises naturally: Are *all* local minima of OPTP I, for large T, approximated in the sense of equations (36)–(38) by the local minima of II? The answer to this question is essentially yes (see comment after Theorem 3). We state the following theorem.

Theorem 3: Let ψ_0 and $\{p_i\}_{i=1,\ldots,N}$ be an initial state and a target population of system (1), respectively. Then, for almost all pairs $(\psi_0, \{p_i\})$ and for large-enough T, the globally optimal solution to the OPTP I is approximated by the globally optimal

solution of II according to Theorem 2. As before, the set of pairs $(\psi_0, \{p_i\})$ of initial states and target populations for which this happens is open and full measure in the corresponding product manifold of initial states $\times$ final populations.

In fact, we prove that, for large-enough T, a number of the lowest energy minima of I that depends (in an unknown way) on T, are approximated by the corresponding lowest energy minima of II according to Theorem 2. However, we think that Theorem 3 is enough to demonstrate the essence of our argument.

Theorem 3 precisely states the main results of our work that were delineated in the introduction: We obtain useful, physically plausible properties of the optimal control and state trajectory, and at the same time, we reduce the solution of the original optimal control problem to a much easier problem. An issue that must be addressed for practical implementation is, how long should the transfer time T be for a reasonable agreement between the first-order approximation of the solution of TPBVP I furnished by the solution of TPBVP II and the actual solution? A proper answer would entail an estimation of the size of the $O(\frac{1}{T^2})$ terms, which is not available. However, we can still provide a useful estimate based on (42) and (43) of Section V (especially the component form). It is straightforward to see that the explicit time dependence on the right-hand sides of (42) and (43) comes from exponentials of the form $e^{i(\omega_{ij}+\omega_{km})t}$ with $(i,j) \neq (m,k)$ and $i \neq j$ or $k \neq m$. Intuitively, the more cycles of the *slowest* varying term take place in time T, the better approximation averaging will be. Hence, a reasonable requirement is

$$\min_{\substack{(i,j)\neq(m,k)\\ i\neq j \text{ or } k\neq m}} |\omega_{ij}+\omega_{km}|\,T \gg 2\pi.$$

IV. Examples

A. Two-State System

Consider the general two-level system ($\psi \in \mathbb{C}^2$) with one control. Then,

$$H_0 = \begin{pmatrix} E_1 & 0 \\ 0 & E_2 \end{pmatrix}, \quad \text{and} \quad V = \begin{pmatrix} V_{11} & V_{12} \\ V_{12}^* & V_{22} \end{pmatrix}.$$

By rescaling u, we make $|V_{12}| = 1$. We are interested in the "population inversion" transfer

$$\psi_0 = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad \longrightarrow \quad \psi_d = \begin{pmatrix} 0 \\ 1 \end{pmatrix}.$$

In this example, the averaged OPTP II can be solved analytically. To begin, we introduce the anti-Hermitian matrices L and $K(L)$ of Section II

$$L = \begin{pmatrix} 0 & L_{12} \\ -L_{12}^* & 0 \end{pmatrix} \quad \text{and} \quad K(L) = L.$$

Then, (29) implies that $L_{12}(s),\ s \in [0,1]$, is constant. Equation (27) reads

$$\frac{d\bar{x}}{ds} = \begin{pmatrix} 0 & L_{12} \\ -L_{12}^* & 0 \end{pmatrix} \bar{x}.$$

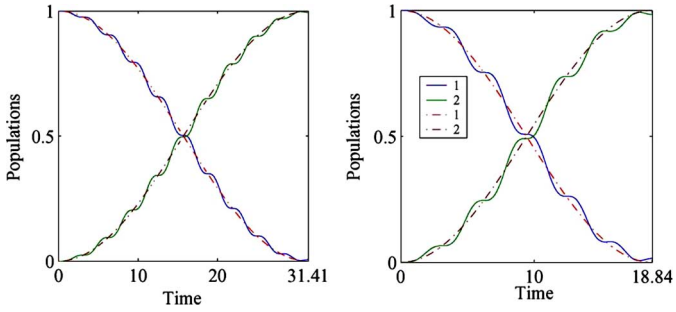

Fig. 1. Averaged (dashed line) and exact (full line) populations of the two-state system under the approximate optimal control, for $T = 10\pi$ and $T = 6\pi$.

Its solution with initial condition $\begin{pmatrix} 1 \\ 0 \end{pmatrix}$ is given by

$$\bar{x}(s) = \begin{pmatrix} \cos(|L_{12}|s) \\ -i\frac{L_{12}}{|L_{12}|}\sin(|L_{12}|s) \end{pmatrix}.$$

To achieve $\bar{x}(1) = \begin{pmatrix} 0 \\ e^{i\beta} \end{pmatrix}$ ($\beta \in [0, 2\pi]$), we must have $|L_{12}| = (n+\frac{1}{2})\,\pi$, with $n \in \mathbb{N}$. The value of the cost (18) is $\frac{\pi^2}{2}(2n+1)^2$. Thus, $n = 0$ corresponds to the global minimum of OPTP II. The approximate optimal control for problem I has the form (38)

$$u(t) = -\frac{\pi}{T}\sin(\omega_{21}t + \varphi)$$

where $\varphi \in [0, 2\pi)$ comes from the phases of L_{12} and V_{12}.

Fig. 1 shows the evolution of populations in a two-state system with $\omega_{21} = 1$ under the approximate optimal control, for $T = 10\pi$ and $T = 6\pi$. Note that even for $T = 6\pi$, the averaged equations are still a good approximation to the full dynamics.

B. Three-State System

Let us now consider the general three-state system with one control. We have,

$$H_0 = \begin{pmatrix} E_1 & 0 & 0 \\ 0 & E_2 & 0 \\ 0 & 0 & E_3 \end{pmatrix}, \quad \text{and} \quad V = \begin{pmatrix} V_{11} & V_{12} & V_{13} \\ V_{12}^* & V_{22} & V_{23} \\ V_{13}^* & V_{23}^* & V_{33} \end{pmatrix}.$$

We assume that $\omega_{12} = E_1 - E_2$, $\omega_{13} = E_1 - E_3$, and $\omega_{23} = E_2 - E_3$ are all different from each other and from zero. Their exact values are unimportant for the averaged OPTP as are the values of V_{11}, V_{22}, and V_{33}. By rescaling the control, we can take $|V_{12}| = 1$ (we assume $|V_{12}| \neq 0$). Define $p \doteq |V_{23}|^2$ and $r = |V_{13}|^2$. We assume that $1 > p > r \geq 0$, with other cases treated similarly. We are interested in the transfer $\psi_0 = (1,0,0)^T \longrightarrow \psi_d = (0,0,1)^T$, particularly in the way the "two-photon" transition $(1,0,0)^T \longrightarrow (0,1,0)^T \longrightarrow (0,0,1)^T$, assists the "direct" ("one-photon") transition $(1,0,0)^T \longrightarrow (0,0,1)^T$.

In this example, we are able to calculate the form of the (locally) optimal controls for the averaged problem analytically up to a constant, which has to be computed by solving the state evolution equations numerically and imposing the terminal conditions on the state. We begin again with (29), $\frac{dL}{ds} = [K(L), L]$.

The diagonal elements of L are zero and L is anti-Hermitian, so it has only three independent (complex) entries, L_{12}, L_{23}, and L_{13}. They satisfy the following equations:

$$\frac{dL_{12}}{ds} = (p-r)L_{13}L_{23}^*$$
$$\frac{dL_{23}}{ds} = (r-1)L_{12}^*L_{13}$$
$$\frac{dL_{13}}{ds} = (1-p)L_{12}L_{23}.$$

Because $\bar{x}_2(0) = \bar{x}_3(0) = 0$, we have that $L_{32}(0) = 0$. The general solution of the aforesaid equations with $L_{32}(0) = 0$ is

$$L_{12}(s) = e^{i\phi_{12}}\, A\mathrm{cn}(ws)$$
$$L_{23}(s) = -e^{i\phi_{23}}\, B\mathrm{sn}(ws)$$
$$L_{13}(s) = e^{i(\phi_{12}+\phi_{23})}\, C\mathrm{dn}(ws)$$

where $w > 0$. cn, sn, and dn are Jacobi elliptic functions and

$$\begin{pmatrix} A \\ B \\ C \end{pmatrix} = \frac{1}{\sqrt{(1-p)(1-r)(p-r)}} \begin{pmatrix} kw\sqrt{p-r} \\ kw\sqrt{1-r} \\ w\sqrt{1-p} \end{pmatrix}.$$

$0 \le k \le 1$ is the modulus of the elliptic functions. Now, from $\bar{x}_1(1) = \bar{x}_2(1) = 0$, we have that $L_{12}(1) = 0$. This allows us to determine w as $w(k) = (2n+1)K(k)$, $n \in \mathbb{N}$, where

$$K(k) \doteq \int_0^{\frac{\pi}{2}} \frac{d\theta}{\sqrt{1-k^2\sin^2\theta}}$$

the complete elliptic integral of the first kind, is the quarter-period of the functions cn and sn. The only undetermined parameter is k. It can be solved for numerically by solving the TPBVP given by

$$\frac{d\bar{x}}{ds} = \begin{pmatrix} 0 & L_{12}(s) & rL_{13}(s) \\ L_{12}^*(s) & 0 & pL_{23}(s) \\ rL_{13}^*(s) & pL_{12}^*(s) & 0 \end{pmatrix} \bar{x}$$

and the boundary conditions

$$\bar{x}(0) = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad \bar{x}(1) = \begin{pmatrix} 0 \\ 0 \\ e^{i\beta} \end{pmatrix}, \quad \beta \in [0, 2\pi].$$

It is straightforward to see that the phases ϕ_{12}, ϕ_{23}, and $\phi_{12} + \phi_{23}$ can be absorbed in the phases of the components of $\bar{x}$, so k is independent of them and depends only on p and r. Intuitively, one expects a discrete set of solutions for k. The expression for the approximate locally optimal controls of OPTP I is

$$u(t) = \frac{2}{T}\Bigg\{ -A\,\mathrm{cn}\left(w(k)\frac{t}{T}\right)\sin(\omega_{21}t - \alpha_{12} + \phi_{12}) + \sqrt{p}\,B\,\mathrm{sn}\left(w(k)\frac{t}{T}\right)\sin(\omega_{32}t - \alpha_{23} + \phi_{23}) - \sqrt{r}\,C\,\mathrm{dn}\left(w(k)\frac{t}{T}\right)\sin(\omega_{31}t - \alpha_{13} + \phi_{12} + \phi_{23})\Bigg\}$$

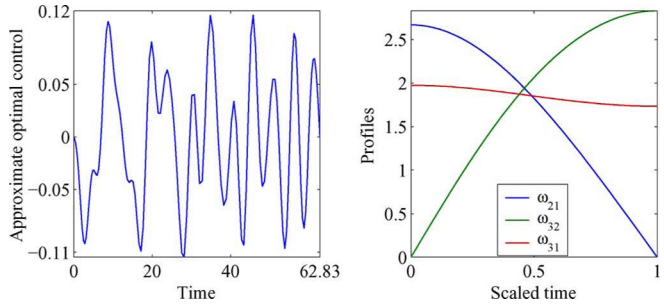

Fig. 2. Approximate optimal control for the transition $1 \to 3$ in $T = 20\pi$ and the corresponding frequency profiles.

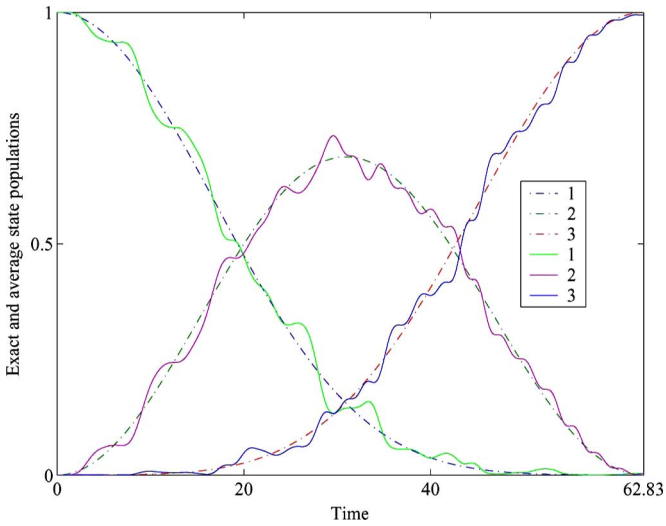

Fig. 3. Averaged (dashed lines) and exact (full lines) populations of the three-state system under the approximate optimal control.

where $\alpha_{ij} = \arg V_{ij}$.

Figs. 2 and 3 show an approximate locally optimal control, the (slowly-varying) profiles of its Bohr frequency components, and the averaged and exact evolution of the three-state system with $p = .9$ and $r = .1$, under this control for $T = 20\pi$. Fig. 3, in particular, demonstrates the point we discussed in the introduction: The "averaged" TPBVP is nonstiff because the short-time-scale natural dynamics of the system has been averaged over, and thus, its solutions are much easier to compute compared with those of the original TPBVP I.

C. Bound States of a Morse Oscillator

Our final example is about the vibrational dynamics of (the ground electronic state of) the OH molecule. A commonly used model for the vibrational dynamics of diatomic molecules is the so-called Morse oscillator. This is an infinite-dimensional model ($\psi \in L^2(\mathbb{R})$), that describes the bounded vibrational as well as the unbounded (relative) motion of the two atoms of the molecule. More details can be found in [14]. Here, we truncate the state space down to the 22-D space of bound states and solve the OPTP II numerically for a transfer from the ground vibrational state (state 1) to an intermediate vibrational state (state 10). Fig. 4 contains the state populations as well as the

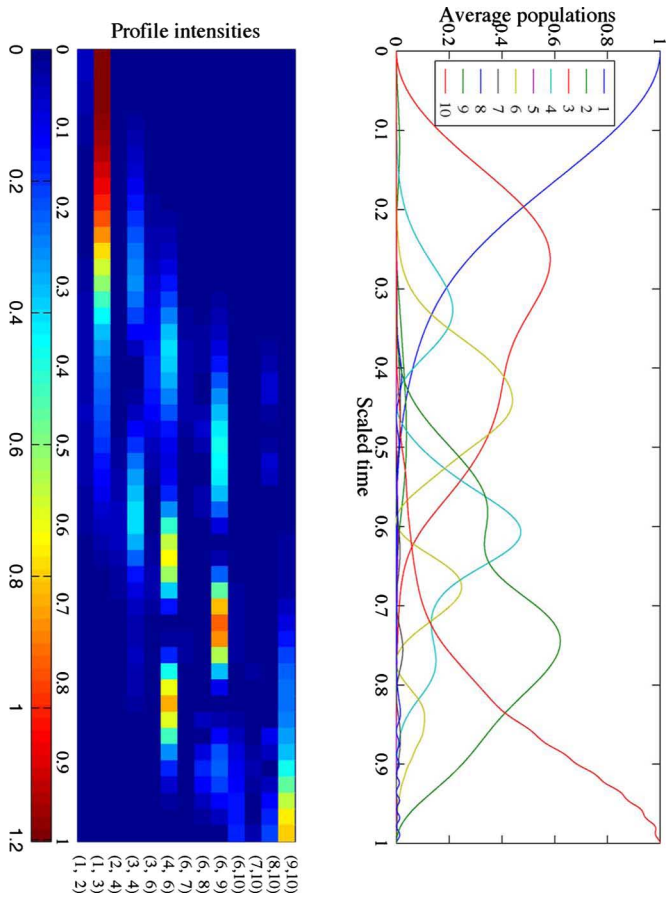

Fig. 4. Average populations and profile intensities versus (scaled) time for a locally optimal transfer $1 \to 10$.

intensities (absolute values squared) of the profiles L_{ij} as functions of the rescaled time. In the pseudocolor plot, the profiles L_{ij} are labeled by the pairs (i, j) of their indices. Note the correspondence between the high-intensity value for a profile L_{ij} and the transition between states i and j.

V. Proof of Main Results

A. Proof of Theorem 1

To begin, we define a new costate variable by $\tilde{\lambda} \doteq T\lambda$ and rewrite (6) and (7) in terms of $\tilde{\lambda}$

$$i\dot{\psi} = H_0\psi + \frac{i}{T}\left(\tilde{\lambda}^* V\psi - \psi^* V\tilde{\lambda}\right) V\psi \tag{39}$$

$$i\dot{\tilde{\lambda}} = H_0\tilde{\lambda} + \frac{i}{T}\left(\tilde{\lambda}^* V\psi - \psi^* V\tilde{\lambda}\right) V\tilde{\lambda}. \tag{40}$$

Note that the form of the boundary conditions remains unchanged, as well

$$\psi(0) = \psi_0, \quad |\psi_i(T)|^2 = p_i, \quad \mathrm{Im}(\psi_i^*(T)\,\tilde{\lambda}_i(T)) = 0. \tag{41}$$

We must show that $e^{-iH_0 t}\,\bar{x}(\frac{t}{T})$ and $e^{-iH_0 t}\,\bar{z}(\frac{t}{T})$ satisfy (39)–(41) up to terms of order $O(\frac{1}{T})$. To this purpose, we perform one more change of variables in (39)–(41)

$$y = e^{iH_0 t}\psi, \quad l = e^{iH_0 t}\tilde{\lambda}.$$

In terms of the new state and costate, the necessary conditions of optimality take the form

$$i\dot{y} = \frac{i}{T}\left(l^* F(t)y - y^* F(t)l\right) F(t)y \tag{42}$$

$$i\dot{l} = \frac{i}{T}\left(l^* F(t)y - y^* F(t)l\right) F(t)l \tag{43}$$

where, as before, $F(t) = e^{iH_0 t}\,V\,e^{-iH_0 t}$, along with

$$y(0) = \psi_0, \quad |y_i(T)|^2 = p_i, \quad \mathrm{Im}(y_i^*(T)\,l_i(T)) = 0. \tag{44}$$

The boundary conditions retain their form because $|y_i(T)|^2 = |\psi_i(T)|^2$ and, $l_i(T)$ and $y_i(T)$ are rotated by the same amount, $e^{-iE_i T}$, with respect to $\psi_i(T)$ and $\tilde{\lambda}_i(T)$. We will refer to (42)–(44) as TPBVP I′. The equivalence of problems I and I′ is evident.

Now, we have to show that $\bar{x}(\frac{t}{T})$ and $\bar{z}(\frac{t}{T})$ satisfy (42)–(44) up to terms of order $O(\frac{1}{T})$. To do this, we average (42) and (43). To make the procedure more transparent, we rewrite (42) and (43) in component form

$$\dot{y}_i = \frac{1}{T}\sum_{km} V_{km}\, e^{i\omega_{km}t}(l_k^* y_m - y_k^* l_m)\sum_j V_{ij}\, e^{i\omega_{ij}t} y_j$$

$$\dot{l}_i = \frac{1}{T}\sum_{km} V_{km}\, e^{i\omega_{km}t}(l_k^* y_m - y_k^* l_m)\sum_j V_{ij}\, e^{i\omega_{ij}t} l_j.$$

One can see (based on our controllability assumption) that we get nonzero contributions from two groups of terms: Terms with $\omega_{km} \neq 0$ and $\omega_{ij} \neq 0$ such that $m = i$ and $k = j$, and terms with $\omega_{km} = \omega_{ij} = 0$, i.e., $k = m$ and $i = j$. Letting $\bar{y}$ and $\bar{l}$ be the averaged y and l, the averaged state and costate equations are

$$\dot{\bar{y}}_i = \frac{1}{T}\left\{\sum_{j\neq i} |V_{ij}|^2(\bar{y}_i\bar{l}_j^* - \bar{l}_i\bar{y}_j^*)\,\bar{y}_j + \sum_k V_{kk}(\bar{y}_k\bar{l}_k^* - \bar{l}_k\bar{y}_k^*)\,V_{ii}\bar{y}_i\right\} \tag{45}$$

$$\dot{\bar{l}}_i = \frac{1}{T}\left\{\sum_{j\neq i} |V_{ij}|^2(\bar{y}_i\bar{l}_j^* - \bar{l}_i\bar{y}_j^*)\,\bar{l}_j + \sum_k V_{kk}(\bar{y}_k\bar{l}_k^* - \bar{l}_k\bar{y}_k^*)\,V_{ii}\bar{l}_i\right\}. \tag{46}$$

To finish the proof, we rescale time in (45) and (46) to $s = \varepsilon t = \frac{t}{T}$. Letting $\tilde{y}(s) \doteq \bar{y}(t)$ and $\tilde{l}(s) \doteq \bar{l}(t)$, these equations read now

$$\frac{d\tilde{y}_i}{ds} = \sum_{j\neq i} |V_{ij}|^2(\tilde{y}_i\tilde{l}_j^* - \tilde{l}_i\tilde{y}_j^*)\,\tilde{y}_j + \sum_k V_{kk}(\tilde{y}_k\tilde{l}_k^* - \tilde{l}_k\tilde{y}_k^*)\,V_{ii}\tilde{y}_i$$

$$\frac{d\tilde{l}_i}{ds} = \sum_{j\neq i} |V_{ij}|^2(\tilde{y}_i\tilde{l}_j^* - \tilde{l}_i\tilde{y}_j^*)\,\tilde{l}_j + \sum_k V_{kk}(\tilde{y}_k\tilde{l}_k^* - \tilde{l}_k\tilde{y}_k^*)\,V_{ii}\tilde{l}_i.$$

These equations are the same as (22) and (23) (with the substitution $\tilde{y} \to \bar{x}$ and $\tilde{l} \to \bar{z}$). Then, from the sequence of variable changes and the averaging theorem, the conclusion of the theorem follows. ■

B. Proof of Theorem 2

To prove Theorem 2, we need the following lemma.

Lemma: Let ψ_0 and $\{p_i\}_{i=1,\dots,N}$ be an initial state and a target population of system (1), respectively, and let $(\bar{x}(s), \bar{z}(s))$ be a solution of TPBVP II over $[0,1]$. The set of pairs $(\psi_0, \{p_i\})$ of initial states and target populations for which all solutions of TPBVP II are *isolated* is open and full measure in the corresponding product manifold.

Proof of Lemma: We begin by introducing new coordinates for the state and costate of the optimal transfer problem II by

$$\bar{x}_i \doteq I_i\, e^{i\phi_i}, \quad \bar{z}_i \doteq J_i\, e^{i\theta_i} \tag{47}$$

where $\phi_i \doteq \arg \bar{x}_i \bmod \pi$ and $I_i \doteq |\bar{x}_i|$ for $\operatorname{Im} \bar{x}_i \geq 0$ and $I_i \doteq -|\bar{x}_i|$ for $\operatorname{Im} \bar{x}_i < 0$, and similarly for the θ_i's and J_i's. Then, $I_i,\ J_i \in \mathbb{R}$ and $\phi_i,\ \theta_i \in [0, \pi)\ \forall i = 1, \dots, N$. With this definition, the phases ϕ_i and θ_i have discontinuities whenever the signs of the imaginary parts of $\bar{x}_i$ and $\bar{z}_i$ change. We shall see that this will not be a problem for us because ϕ_i and θ_i will turn out to be constant in time. On the other hand, the introduction of these coordinates will prove to be beneficial in the following.

In the new coordinates, (31) and (32) take the form

$$\begin{aligned} i\dot{I}_i - I_i\dot{\phi}_i = &+iI_i \sum_{j\neq i} |V_{ij}|^2 I_j J_j\, e^{i(\phi_j - \theta_j)} \\ &- iJ_i\, e^{i(\theta_i - \phi_i)} \sum_{j\neq i} |V_{ij}|^2 I_j^2 \end{aligned} \tag{48}$$

$$\begin{aligned} i\dot{J}_i - J_i\dot{\theta}_i = &-iJ_i \sum_{j\neq i} |V_{ij}|^2 I_j J_j\, e^{i(\theta_j - \phi_j)} \\ &+ iI_i\, e^{i(\phi_i - \theta_i)} \sum_{j\neq i} |V_{ij}|^2 J_j^2 \end{aligned} \tag{49}$$

while the boundary conditions at the end become

$$I_i^2(1) = p_i, \quad I_i(1)J_i(1)\sin(\theta_i(1) - \phi_i(1)) = 0. \tag{50}$$

At this point, we require that $I_i(1) \neq 0$ and $J_i(1) \neq 0\ \forall i = 1, \dots N$. The first set of inequalities for the solution of TPBVP II can be satisfied simply by requiring that $p_i \neq 0$, irrespectively of ψ_0. The second set of inequalities excludes a measure zero set of final populations that depends on ψ_0, though we are not able to give an explicit description of it. Then, the transversality conditions at the endpoint $s = 1$ simplify to

$$\theta_i(1) - \phi_i(1) = 0. \tag{51}$$

The next step is to multiply (48) by J_i, (49) by $-I_i$, add them, and then take the real part of the resulting equation. The outcome of these manipulations is the following equation:

$$\begin{aligned} I_i J_i \frac{d(\phi_i - \theta_i)}{ds} &= \sin(\phi_i - \theta_i) \\ &\times \left\{ I_i^2 \sum_{j\neq i} |V_{ij}|^2 J_j^2 - J_i^2 \sum_{j\neq i} |V_{ij}|^2 I_j^2 \right\}. \end{aligned} \tag{52}$$

For the restricted as earlier, full measure set of final populations, the transversality conditions (51) imply that the solution of (52) satisfies $\theta_i(s) = \phi_i(s)\ \forall s \in [0,1]$. Using this fact, we see that the right sides of (48) and (49) are purely imaginary. This leads to the simplified equations

$$\dot{I}_i = +I_i \sum_{j\neq i} |V_{ij}|^2 I_j J_j - J_i \sum_{j\neq i} |V_{ij}|^2 I_j^2$$

$$\dot{J}_i = -J_i \sum_{j\neq i} |V_{ij}|^2 I_j J_j + I_i \sum_{j\neq i} |V_{ij}|^2 J_j^2$$

$$\dot{\phi}_i(s) = 0, \quad \dot{\theta}_i(s) = 0.$$

We see that the TPBVP II separates nicely into two TPBVPs. First, there is a trivial TPBVP for the phases of state and costate components

$$\dot{\phi}_i(s) = 0, \quad \dot{\theta}_i(s) = 0 \tag{53}$$

$$\phi_i(0) = \phi_{i0}, \ \ \theta_i(1) - \phi_i(1) = 0. \tag{54}$$

This has a unique solution as long as $\psi_{0i} \neq 0,\ \forall i = 1, \dots, N$, so that all $\phi_i(0)$ are unambiguously defined. Then, there is the TPBVP for the (signed) magnitudes of state and costate components

$$\dot{I}_i = +I_i \sum_{j\neq i} |V_{ij}|^2 I_j J_j - J_i \sum_{j\neq i} |V_{ij}|^2 I_j^2 \tag{55}$$

$$\dot{J}_i = -J_i \sum_{j\neq i} |V_{ij}|^2 I_j J_j + I_i \sum_{j\neq i} |V_{ij}|^2 J_j^2 \tag{56}$$

$$I_i(0) = I_{i0}, \quad I_i^2(1) = p_i. \tag{57}$$

We will refer to this as the *real form* of problem II. This TPBVP can be seen to express the necessary conditions for optimal population transfers of the following *real* control system on S^{N-1}:

$$\frac{dI}{ds} = \begin{pmatrix} 0 & |V_{12}|v_{12} & \cdots \\ -|V_{21}|v_{21} & 0 & \cdots \\ \vdots & \vdots & \ddots \end{pmatrix} I. \tag{58}$$

with cost functional similar to that of OPTP II, namely

$$\int_0^1 \sum_{i\neq j} v_{ij}^2(s)\, ds.$$

Here the v_{ij} are real controls and (58) is controllable on S^{N-1} because of the controllability assumption on the original system (see the Appendix). This separation of the TPBVP II into a trivial problem for the evolution of the arguments of state and costate components and real TPBVP II was inspired by [7, Section 4]. Recall that, to effect this reduction of TPBVP II to its real form, we had to exclude a subset of pairs $(\psi_0, \{p_i\})$ of initial states

and target populations. This subset is of measure zero in the product manifold of initial states $\times$ final populations.

We are now ready to prove the lemma. Fix an initial condition $I(0)$. We define a function $\mathcal{G}:\mathbb{R}^{N-1}\to S^{N-1}$ of the initial costate vector $J(0)$ of real problem II by

$$\mathcal{G}(J(0)) \doteq I(1). \tag{59}$$

$\mathcal{G}$ is a function of only $N-1$ variables because $J(0)$ is a tangent vector to S^{N-1} at $I(0)$, and hence, it is always perpendicular to $I(0)$. Another way to see this is from the condition $0=\bar{z}^*(0)\bar{x}(0)=J^T(0)I(0)$, which comes from the identity $\bar{z}^*(s)\bar{x}(s)=0$, at $s=0$. This fixes one of the components of $J(0)$, say $J_1(0)$, in terms of the others. In the following, when talking about initial costate vectors, we will identify vectors in $\mathbb{R}^{N-1}$ with vectors in $\mathbb{R}^N$ perpendicular to $I(0)$. We explore now some of the properties of $\mathcal{G}$. First, it is a smooth function of its arguments. Indeed, the right sides of (55) and (56) are C^∞ functions of I and J, so their solutions depend smoothly on $I(0)$ and $J(0)$, according to the differentiability theorem for solutions of ordinary differential equations [15, Section 32]. Thus, when $I(0)$ is fixed, $\mathcal{G}(J(0))\doteq I(1)$ depends smoothly on $J(0)$. Second, the controllability of system (58) guarantees that $\mathcal{G}$ is onto. However, $\mathcal{G}$ is a mapping between manifolds of the same dimension. From these, we can conclude the following:

1) Sard's theorem [16] guarantees that the set of critical values of $\mathcal{G}$ in S^{N-1} is of measure zero, or equivalently, that the set of regular values of $\mathcal{G}$ in S^{N-1} is of full measure.
2) $\mathcal{G}$ is a local submersion from (open) neighborhoods of $\mathbb{R}^{N-1}$ to neighborhoods of S^{N-1}, and furthermore, a local diffeomorphism between such neighborhoods.
3) The set of regular values of $\mathcal{G}$ in S^{N-1} is open and is of full measure.

Now, fix such a regular value I_1 of $\mathcal{G}$ in S^{N-1}. Due to the local diffeomorphism property, the preimage $\mathcal{G}^{-1}(I_1)$ must be a discrete set of points. Also, it cannot have a limit point. Indeed, if there exists a subsequence $\{J_0\}_{n_k}\to\hat{J}_0$ such that

$$\mathcal{G}(\hat{J}_0)=\lim_{k\to\infty}\mathcal{G}(\{J_0\}_{n_k})=I_1$$

then it is easy to see that the Jacobian of $\mathcal{G}$ at $\hat{J}_0$ is not full rank (equivalently its null space $\mathcal{N}(D\mathcal{G}(\hat{J}_0))\neq\emptyset$). This, however, would violate the assumption that $I_1=\mathcal{G}(\hat{J}_0)$ is a regular value of $\mathcal{G}$. Hence, given an initial state $I(0)$ and a final state $I(1)$ (satisfying all the requirements we have discussed so far), the set of initial costates $J(0)$ that achieves the required transfer (for the real form of TPBVP II) is a set of discrete, isolated vectors. This conclusion still holds if we consider given final populations p_i, because there is a finite number of final states $I(1)$ such that $I_i^2(1)=p_i$ (there are 2^N such states $I(1)$, where N is the dimension of the original system, since all $I_i(1)\neq 0$). Finally, since the TPBVP for the phases of state and costate components has a unique solution (given the restrictions on ψ_0 and the p_i's), the conclusion holds for TPBVP II as well. Hence, given an initial state, the set of populations that can be achieved by *isolated* locally optimal transfers of TPBVP II is open and full measure. Then, the set of pairs $(\psi_0,\{p_i\})$ of initial states and target populations for which all solutions of TPBVP II are *isolated* is open and in full measure in the corresponding product manifold. ■

Proof of Theorem 2: We are going to define functions $\mathcal{F}$ and $\mathcal{F}_1$ for TPBVPs II and I′ analogous to $\mathcal{G}$ for the real form of TPBVP II used in the proof of the lemma. First, for TPBVP II, we define $\mathcal{F}:\mathbb{R}^{2N-2}\longrightarrow\mathbb{R}^{2N-2}$ by

$$\mathcal{F}(\bar{z}(0)) \doteq (|\bar{x}_2^2(1)|,\ldots,|\bar{x}_N^2(1)| \\ \mathrm{Im}(\bar{x}_2^*(1)\,\bar{z}_2(1)),\ldots,\mathrm{Im}(\bar{x}_N^*(1)\,\bar{z}_N(1)))^T.$$

Recall that one complex component of $\bar{z}(0)$ is fixed by $\bar{x}^*(0)\bar{z}(0)=0$. Here, we again identify initial costate vectors in $\mathbb{C}^N$ perpendicular to $\bar{x}(0)=\psi_0$ with vectors in $\mathbb{C}^{N-1}\sim\mathbb{R}^{2N-2}$. $\mathcal{F}$ is a smooth function of its arguments. Also, any initial costate vector $\bar{z}(0)$ such that $\mathcal{F}(\bar{z}(0))=(p_2,\ldots,p_N,0,\ldots,0)^T$ provides a solution to problem II.

The corresponding terminal condition function $\mathcal{F}_1$ for problem I′ is defined exactly the same way

$$\mathcal{F}_1(l(0);T) \doteq (|y_2(T)|^2,\ldots,|y_N(T)|^2 \\ \mathrm{Im}(y_2^*(T)\,l_2(T)),\ldots,\mathrm{Im}(y_N^*(T)\,l_N(T)))^T.$$

The second argument of $\mathcal{F}_1$ is just a reminder of the transfer time. $\mathcal{F}_1$ is also a smooth function of its arguments. Again, any initial costate vector $y(0)$ such that $\mathcal{F}_1(y(0))=(p_2,\ldots,p_N,0,\ldots,0)^T$ provides a solution to problem I'. From the proof of Theorem I, we know that

$$\mathcal{F}_1(v;T)=\mathcal{F}(v)+O\left(\frac{1}{T}\right). \tag{60}$$

From this, we also have that

$$D_v\mathcal{F}_1(v;T)=D\mathcal{F}(v)+O\left(\frac{1}{T}\right).$$

Although $\mathcal{F}_1$ is not formally defined for $T=\infty$, we can define it from (60) as $\mathcal{F}_1(v;\infty)\doteq\mathcal{F}(v)$. With this definition, $\mathcal{F}_1$ is continuous in $\frac{1}{T}$ at $\frac{1}{T}=0$, with continuous first derivatives in v and $\frac{1}{T}$ there. In particular, $D_v\mathcal{F}_1(v;\infty)=D\mathcal{F}(v)$.

Consider now an initial costate vector v as follows that solves problem II

$$\mathcal{F}(v)=(p_2,\ldots,p_N,0,\ldots,0)^T.$$

We have seen that the set of pairs $(\psi_0,\{p_i\})$ such that *all* v's that satisfy this are isolated ($D\mathcal{F}(v)$ full rank), is an open set of full measure in the product space of (initial states) $\times$ (final populations). For such a transfer and for large-enough T, the implicit function theorem guarantees the existence of a δv such that $\mathcal{F}_1(v+\delta v;T)=(p_2,\ldots,p_N,0,\ldots,0)^T$. Then, $v+\delta v$ provides a solution for problem I′ and $\frac{v+\delta v}{T}$ is a solution for problem I. Taking T large enough so that $\|\delta v\|=O(\frac{1}{T})$, the averaging theorem guarantees that this solution $(\psi(t),\lambda(t))$ to TPBVP I is such that

$$\psi(t)=e^{-iH_0t}\,\bar{x}\left(\frac{t}{T}\right)+O\left(\frac{1}{T}\right)$$

$$\lambda(t)=\frac{1}{T}e^{-iH_0t}\,\bar{z}\left(\frac{t}{T}\right)+O\left(\frac{1}{T^2}\right)$$

where $(\bar{x}(s),\bar{z}(s))$ is the solution to problem II we started with. ■

C. Proof of Theorem 3

We only consider transfers such that problem II has isolated solutions. Any initial costate v that satisfies $\mathcal{F}(v) = (p_2, \ldots, p_N, 0, \ldots, 0)^T$ provides a solution to problem II and $D\mathcal{F}(v)$ is full rank. Inside a ball of radius $M > 0$, there can only be a *finite* number of these initial costates v because any discrete set with no limit points inside a compact set must be finite.

From (60), we may conclude that, for a given transfer, we can take T large enough to bound the difference of $\mathcal{F}$ and $\mathcal{F}_1$ over an open ball around the origin by any $\delta > 0$: $\forall\, M > 0\, \forall\, \delta > 0,\ \exists\, T_* > 0$ such that for $T > T_*$, $|\mathcal{F}_1(v;T) - \mathcal{F}(v)| < \delta\ \forall\, |v| < M$. Thus, the only solutions of $\mathcal{F}_1(\tilde{v};T) = (p_2, \ldots, p_N, 0, \ldots, 0)^T$ inside the ball of radius M come from perturbing solutions of $\mathcal{F}(v) = (p_2, \ldots, p_N, 0, \ldots, 0)^T$ by quantities of order $O(\frac{1}{T})$, for T large enough. In particular, they also form a finite set and $D_v\mathcal{F}_1(\tilde{v};T)$ is full rank for each such $\tilde{v}$. We arrive at exactly the same conclusion if we consider open ellipsoids (rather than balls) around the origin.

We introduce now the following quadratic form in $\{v \in \mathbb{C}^N \text{ s.t. } \psi_0^* v = 0\} \simeq \mathbb{C}^{N-1} \simeq \mathbb{R}^{2N-2}$

$$E(v) = \sum_{i \neq j}^{N} |V_{ij}|^2 |\psi_{0i} v_j^* - v_i \psi_{0j}^*|^2.$$

E is nonnegative, and furthermore, due to the connectivity of the graph of V (part of the controllability assumption) can be shown to be positive definite. Indeed, suppose that $\exists v \neq 0,\ \psi_0^* v = 0$, such that $E(v) = 0$. Then, we have that

$$\psi_{0i} v_j^* - v_i \psi_{0j}^* = 0, \quad \forall\, (i,j) \text{ s.t. } V_{ij} \neq 0. \tag{61}$$

Let us assume for a moment that $V_{12} \neq 0$. Then (recall, all $\psi_{0i} \neq 0$)

$$\frac{v_2}{\psi_{02}} = \frac{v_1^*}{\psi_{01}^*}.$$

It is straightforward to see that the connectivity of the graph of V and the repeated use of the relations (61) allows us to show that

$$\frac{v_i}{\psi_{0i}} = \frac{v_1}{\psi_{01}} \quad \text{or} \quad \frac{v_1^*}{\psi_{01}^*} \quad \forall\, i = 2, \ldots, N.$$

Recall now that both ψ_0 and v can be defined modulo global phases that we choose such that ψ_{01} and v_1 are real. Then,

$$\frac{v_i}{\psi_{0i}} = \frac{v_1}{\psi_{01}}, \forall\, i = 2, \ldots, N.$$

The relation $\psi_0^* v = 0$ implies that

$$\left(\sum_{i=1}^{N} |\psi_{0i}|^2 \right) \frac{v_1}{\psi_{01}} = 0$$

which means that $v_1 = 0$, and hence, $v = 0$.

Since E is positive definite, its sublevel sets define ellipsoids in $\mathbb{R}^{2N-2}$. The significance of our choice for $E(v)$ lies in the following:

$$\bar{H}(\bar{x}, \bar{z}) = \sum_{i \neq j} |V_{ji}|^2 |\bar{x}_i \bar{z}_j^* - \bar{z}_i \bar{x}_j^*|^2$$

is the Hamiltonian function from which the optimal state and costate (31) and (32) are derived. Thus, $\bar{H}$ is a constant of motion along the optimal solutions. Note also that $\bar{H}(\bar{x}, \bar{z}) = \sum_{i \neq j} |u_{ij}|^2$. Since E is just $\bar{H}$ evaluated at $t = 0$, we conclude that E is equal to the cost of a trajectory of system II [(31) and (32)] with initial conditions (ψ_0, v)

$$\int_0^1 \sum_{i \neq j} |u_{ij}(s)|^2 ds = \sum_{i \neq j}^{N} |V_{ij}|^2 |\psi_{0i} v_j^* - v_i \psi_{0j}^*|^2.$$

With a calculation similar to that of Section II, one can show that the locally optimal costs for problems II and I/I′ coming from the solutions v and $\tilde{v} = v + O(\frac{1}{T})$, respectively, are related as follows:

$$\int_0^T u^2(t)\, dt = \frac{1}{T} \left(\int_0^1 \sum_{i \neq j} |u_{ij}(s)|^2\, ds + O\left(\frac{1}{T}\right) \right)$$
$$= \frac{1}{T} \sum_{i \neq j}^{N} |V_{ij}|^2 |\psi_{0i} v_j^* - v_i \psi_{0j}^*|^2 + O\left(\frac{1}{T^2}\right).$$

Let us fix a value $E_0 > 0$ such that the initial costate v_0 that achieves the desired transfer with the *minimum* energy for TPBVP II satisfies

$$\sum_{i \neq j}^{N} |V_{ij}|^2 |\psi_{0i} v_j^* - v_i \psi_{0j}^*|^2 < E_0.$$

Then, for large-enough T, $\tilde{v}_0 = v_0 + O(\frac{1}{T})$ is the initial costate that achieves the desired transfer with the *minimum* energy for problem I′. This proves the assertion of the theorem. In fact, we proved a little bit more: Not only the global optimum, but also all local optima of problem I′ with values of energy less than $\frac{E_0}{T}$, come from local optima of problem II according to Theorem 2, for T large enough (Note that the solutions $\tilde{v}$ of problem (I/I′) *outside* the ellipsoid $E(\tilde{v}) < E_0$ have higher costs than those inside the ellipsoid). ■

VI. Conclusion

We examined the large transfer time limit of exact, optimal population transfers in a finite-dimensional quantum system. The investigation of this problem uncovered useful structure in the optimal control and state trajectory, and moreover, resulted in the much simpler optimal control problem II whose solution provides the first-order solution to the original optimal transfer problem, in a $\frac{1}{T}$ expansion. The main reason we considered *exact* population transfers is that in this case, the "average" TPBVP II (22)–(24) is independent of the transfer time T of the original problem. This, in turn, allowed us to prove Theorems 2 and 3 that are important for uncovering the structure of solutions of OPTP I. On the other hand, solving TPBVP II itself becomes more and more difficult as the dimension of the system grows. For practical applications, it is preferable to solve population transfer problems that do not require *exact* terminal populations but, rather, attempt to minimize a function of the terminal populations. Such a function should be selected so that at its minimum the final populations are close to a desired

distribution. This leads to OPTP with *relaxed* cost functionals such as

$$a\int_0^T u^2(t)\,dt+\sum_{i=1}^{N}\left(|\psi_i(T)|^2-p_i\right)^2 \text{ or}$$

$$a\int_0^T u^2(t)\,dt+\sum_{i=1}^{N} m_i|\psi_i(T)|^2$$

where $a>0$ is a weight factor on the control effort (energy). The advantage of OPTPs like these is that they lead to TPBVPs with *separated* boundary conditions that are amenable to converging, iterative solution techniques involving numerical solutions of initial value problems only [17]. We think that the methods used in this paper, namely the averaged control system (17) and the averaged OPTP II, could be used successfully for these problems, too. Preliminary investigations show that the analog of Theorem 1 still holds, although we have not been able to prove the analogs of Theorems 2 and 3 yet. The computational advantage brought by the use of averaged TPBVPs in these problems is great and good approximate solutions of the above OPTPs can be computed with low computational expense, compared to the full solution. We hope to report more on these matters in a future publication.

APPENDIX

In this appendix, we prove various facts stated in the paper. First, we prove the transversality conditions $\mathrm{Im}(\psi_i^*(T)\,\lambda_i(T))=0,\ i=1,\dots,N$. The OPTP I is a standard Bolza problem with (real) terminal state constraints, $|\psi_i(T)|^2=p_i,\ i=1,\dots,N$. According to the general theory of such problems [18], the transversality conditions for the costate at the final time are given by

$$\lambda_i(T)=\nu_i\,\frac{\partial(|\psi_i(T)|^2-p_i)}{\partial\psi_i(T)^*}=\nu_i\psi_i(T) \tag{62}$$

where ν_i are the real Lagrange multipliers that enforce the terminal state constraints. The transversality conditions in the form we state them

$$\mathrm{Im}(\psi_i^*(T)\,\lambda_i(T)\,)=0,\quad i=1,\dots,N$$

follow easily.

Next, we prove that $\lambda(t)^*\psi(t)=0$. One can easily show from (3) and (4) that $\lambda(t)^*\psi(t)$ is constant along any optimal trajectory and so, $\lambda(t)^*\psi(t)=\lambda(T)^*\psi(T)$. From the transversality conditions, it follows that $\mathrm{Im}(\psi^*(T)\,\lambda(T))=0$, and thus, $\mathrm{Im}(\psi^*(t)\,\lambda(t))=0$. Let us now decompose λ as follows, $\lambda=c\psi+\lambda_\perp$, with c real and $\lambda_\perp$ perpendicular to ψ, i.e., $\lambda_\perp^*\psi=0$. All we need to show is that $c=0$. We introduce this representation of λ into (5). The resulting equation is

$$u=i\,(\lambda_\perp^*V\psi-\psi^*V\lambda_\perp)+i\,(c^*-c)\,(\psi^*V\psi)$$

and the reality of u forces c to be zero.

Finally, we prove that system (17) is controllable on account of the controllability assumption on the original system (1). Denote by E_{ij} the matrices with only one nonzero element equal to one, at the position (i,j). So, $(E_{ij})_{kl}=\delta_{ik}\delta_{jl}$. It is easy to see that $E_{ij}E_{mn}=\delta_{jm}E_{in}$. Due to the fact that (17) has complex controls, every nonzero $V_{ij}, i\neq j$, provides us with two generators of $su(N)$ (traceless anti-Hermitian matrices), $E_{ij}-E_{ji}$ and $i(E_{ij}+E_{ji})$. One can easily verify the following commutation relations:

$$[E_{ij}-E_{ji},E_{jk}-E_{kj}]=E_{ik}-E_{ki} \tag{63}$$

$$[E_{ij}-E_{ji},i(E_{jk}+E_{kj})]=i(E_{ik}+E_{ki}) \tag{64}$$

$$[E_{ij}-E_{ji},i(E_{ji}+E_{ij})]=2i(E_{ii}-E_{jj}) \tag{65}$$

where $i\neq k$. Due to the connectivity of the graph of V, there exists a sequence of index pairs that connects any state index i with any other state index j. Thus, starting with the given generators $E_{ij}-E_{ji}$ and $i(E_{ij}+E_{ji})$ for all $V_{ij}\neq 0, i\neq j$, we can generate, with repeated use of (63) and (64), all missing such generators (corresponding to $V_{ij}=0, i\neq j$). Finally, using (65), we can generate the diagonal generators of $su(N)$ (its Cartan subalgebra). For the system (58), which has real controls, every nonzero $V_{ij}, i\neq j$, provides us with a generator of $so(N)$ (antisymmetric matrices), $E_{ij}-E_{ji}$. In that case, the repeated use of (63) is enough to establish controllability.

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