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Optimal population transfers for a quantum system in the limit of large transfer time

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***Abstract*— We consider an optimal population transfer problem for a finite-dimensional quantum system with an energy-like cost. We show that a way to realize a small control limit is as the limit of large transfer time T. In the process we show that, in the large T limit, the optimal control is a sum of terms with the following structure: Each term is an exponential with frequency given by a Bohr frequency of the quantum system times a slow varying envelope, that is a function of $\frac{t}{T}$. The form of these envelopes can be computed by solving an "averaged" two-point boundary value problem. We demonstrate our results with an example.**

I. Motivation and background

One of the most fundamental and practical problems in Chemistry is to influence the outcome of chemical reactions. Traditionally, one would do this by adjusting environmental variables (temperature, pressure) and by inserting various catalysts in the reacting system. Although highly successful in many cases, this approach still leaves much to be desired. A modern approach is to use pulses of laser light to break existing chemical bonds in molecules so that new ones may be created. If successful, this would be a much more precise way to achieve desired outcomes of reactions.

For this program to be successful, one needs to control the dynamics of reacting atoms and molecules at the microscopic level. In this regime, one needs the language of Quantum Mechanics for a correct description of the dynamics. Understanding how to control this dynamics under the limitations posed by each individual quantum system is a challenging task.

Before we formulate the problem of interest, we begin with a brief description of quantum systems. The state space of a quantum system is an appropriate, finite or infinite-dimensional complex Hilbert space $\mathcal{H}$. The evolution law is Schrödinger's equation:

$$i\hbar\,\dot{\psi} = H_0\psi, \tag{1}$$

where H_0 is a self-adjoint operator in $\mathcal{H}$, referred to as the system Hamiltonian, $\psi \in \mathcal{H}$ is the state variable and $\hbar$ is Planck's constant divided by 2π. This evolution law is linear and unitary (norm preserving). By convention, we normalize $\|\psi\| = 1$. This implies that the actual state space of the system is the sphere $\|\psi\| = 1$ in $\mathcal{H}$. Also, the physical interpretation of the quantum mechanical formalism (which we are not going to go into) requires that we identify states that differ only by a total phase factor, $\psi \sim e^{i\phi}\psi$.

The (normalized) eigenvectors and eigenvalues of H_0,

$$H_0 e_i = E_i e_i,\ i = 1, \ldots, N$$

(N is the dimension of $\mathcal{H}$), are of great physical importance. Indeed, the E_i's are the possible energies of the system and the energy "eigenstates" e_i, are the possible states at which one can find the system after measuring its energy. If we expand a state vector ψ in the orthonormal basis furnished by the e_i's,

$$\psi = \sum_{i=1}^{N} \psi_i e_i,$$

the quantities $|\psi_i|^2 = |e_i'\psi|^2$ are called the populations of the energy states. The solution of (1) is

$$\psi(t) = e^{-iH_0 t/\hbar}\,\psi(0) \ = \sum_{i=1}^{N} \psi_i(0)\, e^{-iE_i t/\hbar}\, e_i.$$

Note that the dynamics preserves the populations of the energy states, because

$$|\psi_i(t)|^2 = |\psi_i(0)\, e^{-iE_i t/\hbar}|^2 = |\psi_i(0)|^2.$$

In particular, if $\psi(0) = e_i$, then $\psi(t) = e^{-iE_i t}\, e_i$ and so the energy eigenstates are equilibria of the dynamics.

When a quantum system is acted upon by an external field (e.g. an atom in the electric field of a laser), (1) is modified:

$$i\hbar\,\dot{\psi} = (\, H_0 + \sum_{\alpha} H_\alpha u_\alpha(t)\,)\,\psi. \tag{2}$$

The H_α's are referred to as control Hamiltonians and describe the coupling of the system to the external fields $u_\alpha(t)$. System (2) is controllable if, given any initial state ψ_0 and any target state ψ_d (both defined modulo a total phase), there exists a transfer time T and control functions $u_\alpha : [0, T] \to \mathbb{R}$ such that, the solution of (2) with $\psi(0) = \psi_0$ satisfies $\psi(T) = \psi_d$ (both ψ_0 and ψ_d are defined modulo phases). The controllability analysis of (2) for finite dimensional quantum systems is based on controllability results for right invariant systems on Lie groups ([1], [2], [3]). We quote the main result and refer to the bibliography for more details: The system (2) is controllable if the Lie Algebra generated by $-iH_0$ and the $-iH_\alpha$'s is $su(N)$, the Algebra of traceless anti-symmetric complex $N \times N$ matrices.

The design of controls that steer a quantum system to desired target states is a very important problem, see [4], [5], [6] for constructive approaches, [7], [8], [9], [10] for optimal control approaches based on energy-like objectives, [11], [12] for Lyapunov-based approaches and [13] for time-optimal control. We would like to differentiate between *co-herent* state transfers, where the *relative* phases between the

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components of the final state are important and *incoherent* state transfers, where only a final population is desired and the relative phases between the components of the final state are not important. It is the importance of incoherent state transfers to many applications in Quantum Chemistry, that motivates our study of the structure of optimal controls for such transfers.

Before we get into more detail, we would like to mention an important concept from Quantum Mechanics, which will prove to be significant in the development. This is the notion of Bohr frequencies of a quantum system: For every pair of eigenstates, e_i and e_j, the quantity

$$\omega_{ij} = \frac{E_i - E_j}{\hbar},$$

(or rather its absolute value) is referred to as the Bohr frequency of the transition between states i and j. The reason is the following: If one tunes the external field exactly to this frequency, one can transfer all the population of state i to state j (and vice versa) in a sufficiently long time. Let's be more precise. For simplicity, consider a quantum system with one control (which is usually the case) and set $\hbar$ to 1:

$$i\dot{\psi} = (H_0 + H_1 u(t))\psi. \tag{3}$$

One can show with an averaging argument that, using the control $u(t) = \frac{\pi}{|H_{1ij}|T}\cos(\omega_{ij}t + \phi)$ for a large time T, results to the following final populations:

$$\begin{aligned} |\psi_i(T)|^2 &= |\psi_j(0)|^2 + O(\frac{1}{T}), \\ |\psi_j(T)|^2 &= |\psi_i(0)|^2 + O(\frac{1}{T}), \\ |\psi_k(T)|^2 &= |\psi_k(0)|^2 + O(\frac{1}{T}),\ k \neq i, j. \end{aligned}$$

So, the Bohr frequencies are resonance frequencies for the interaction of a quantum system with external fields. In many applications in Physics and Chemistry, one wants to cause a transition from a fully populated energy eigenstate to an unpopulated one. For this, a field of the right frequency will suffice. If it happens that the matrix element of the control Hamiltonian between these two states is zero or very small, one would have to use transitions through intermediate states. That is, one would have to mix and time fields with the right Bohr frequencies. So far, intuition has been the only guide on how to do this.

If we consider the more general problem of transfering an initial state to a target *distribution* of populations using fields of the right Bohr frequencies and timing, intuition can hardly be of any help. This is even more so if we would like this transfer to be optimal in some sense, for example in terms of energy used to achieve it.

Here, we consider the general problem of optimizing a population transfer with respect to the control energy. We show that for large transfer times, the previous picture emerges naturally: The optimal field is a sum of terms, with each term being an exponential with a Bohr frequency multiplied by a slow envelope, i.e. a function of $\frac{t}{T}$. These envelopes control the timing and mixing of the various frequencies. We also show how to compute these envelopes by solving an "averaged" two-point boundary value problem.

Before we leave this introduction, we would like to comment on two issues. First, regarding the choice of optimal control design in contrast to other design techniques, like those mentioned above. As far as constructive techniques are concerned, the calculations involved in the design become impossible as the size of the system increases and typically, the system size in Quantum Chemistry can be very large. This is not to say of course that the difficulty of solving the optimal control problem does not increase as the system size increases.

We would also like to comment on the important issue of how big the transfer time can be. For many applications, especially to Quantum Computing, transfer times must be short. The main reason for this is that (2) does not take into consideration the effects of the environment on the quantum system and so, any control design based on it would be invalidated for times of the order of the so called decoherence time. In cases like these one should use other control strategies, e.g. time-optimal control [13].

For Quantum Chemistry, the time constraints come from the fact that, in a molecule, besides the vibrational dynamics which is of interest in dissociation phenomena, there is also rotational and electronic dynamics. One would like to achieve dissociation in a time scale where the other dynamics is not important. It turns out that in many cases, this time scale is large enough for our approximation to be useful.

II. Optimal population transfers

In this approach to control design, one singles out controls that achieve a desired transfer and, in addition, optimize a certain performance index. The energy-like performance index

$$J = \frac{1}{2}\int_0^T u^2(t)\,dt,$$

has a physical appeal and leads to interesting conclusions. We pose the following problem: Find a $u(t), t \in [0, T]$, that minimizes J and drives an initial state ψ_0 of system (3) to a target population $\{|\psi_i(T)|^2 = p_i\}_{i=1,\ldots,N}$.

The Maximum Principle of optimal control provides necessary conditions for optimality in terms of the Hamiltonian function

$$H(\psi, \lambda, u) = \frac{1}{2}u^2 - i\lambda'(H_0 + H_1 u)\psi + i\psi'(H_0 + H_1)\lambda, \tag{4}$$

where λ is the co-state vector. The optimal control u and the corresponding state and co-state 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

$$\begin{aligned} i\dot{\psi} &= (H_0 + H_1 u)\psi, & (5)\\ i\dot{\lambda} &= (H_0 + H_1 u)\lambda, & (6)\\ u &= i(\lambda' H_1 \psi - \psi' H_1 \lambda). & (7) \end{aligned}$$

To these equations one must append the boundary conditions

$$|\psi_0'\psi(0)|^2 = 1, \quad |\psi_i(T)|^2 = p_i \tag{8}$$

and

$$\mathrm{Im}(\psi_0'\lambda(0)) = \mathrm{Im}(\psi_i^*(T)\lambda_i(T)) = 0, \tag{9}$$

$\forall\ i = 1,\ldots,N$ (transversality conditions). Also, λ is perpendicular to ψ, that is $\lambda'(t)\psi(t) = 0,\ t \in [0,T]$. These equations define a two-point boundary value problem which has proved to be very hard to solve, both analytically and numerically. On the analytical side, only a few special cases have been solved: Transfers between energy eigenstates in two- and three-state systems with the so-called $k+p$ structure [14]. Also, a semi-analytical solution exists for the case of a two-state system with one control only, where the functional form of the control can be determined analytically, but the two-point boundary value problem can be solved only numerically [8]. We shall comment on the difficulty of the numerical solution of this problem in section VI.

III. An approximation based on averaging

In this section, we present an approximation to the two-point boundary value problem (5) - (9) in the limit of small control amplitude. In the next section we show that this limit can be realized as a large transfer time limit and so, we end up with an approximate form for the optimal control in the large T limit.

To begin, we change variables (transform to the "Interaction Picture"),

$$\begin{aligned} \psi &= e^{-iH_0t}\chi, \\ \lambda &= e^{-iH_0t}\zeta. \end{aligned}$$

The time evolution of the new variables χ and ζ is due entirely to the control, because the free evolution has been accounted for. In the new variables, the necessary conditions of optimality take the form

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

$$i\dot{\zeta} = u\,F(t)\zeta, \tag{11}$$

$$\begin{aligned} u &= i\,(\zeta' F(t)\chi - \chi' F(t)\zeta) \\ &= i\,\mathrm{tr}[F(t)(\chi\zeta' - \zeta\chi')], \end{aligned} \tag{12}$$

where $F(t) = e^{iH_0t}\,H_1\,e^{-iH_0t}$, along with

$$|\psi_0'\chi(0)|^2 = 1, \quad |\chi_i(T)|^2 = p_i \tag{13}$$

and

$$\mathrm{Im}(\psi_0'\zeta(0)) = \mathrm{Im}(\chi_i^*(T)\zeta_i(T)) = 0. \tag{14}$$

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

$$F_{ij}(t) = H_{1ij}e^{i(E_i-E_j)t} = H_{1ij}e^{i\omega_{ij}t}.$$

We now define a normalized co-state variable $\hat{\zeta}$, by $\zeta(t) = \varepsilon\hat{\zeta}(t)$, where $\varepsilon = \|\zeta\|$. Using this new variable, we rewrite equations (10), (11) and (12) using u from (12):

$$\dot{\chi} = \varepsilon\,(\hat{\zeta}' F(t)\chi - \chi' F(t)\hat{\zeta})\,F(t)\,\chi, \tag{15}$$

$$\dot{\hat{\zeta}} = \varepsilon\,(\hat{\zeta}' F(t)\chi - \chi' F(t)\hat{\zeta})\,F(t)\,\hat{\zeta}, \tag{16}$$

$$u = i\,\varepsilon\,\mathrm{tr}[F(t)(\chi\hat{\zeta}' - \hat{\zeta}\chi')]. \tag{17}$$

From (17), we see that ε controls the "size" of u (in fact, since χ and $\hat{\zeta}$ are normalized, the amplitude of u is less or equal to $2\varepsilon\|H_1\|$). We want to consider the limit of small control amplitude, i.e. $\varepsilon \ll 1$. Note that the determination of ε is part of the two-point boundary value problem so, we cannot make any a priori statements about its size. We'll see in the next section how the small ε limit can be realized. For now, we want to approximate equations (15) - (17) for small ε.

The ideal approximation method in our case is averaging. In averaging, one deals with equations of the form

$$\dot{y} = \varepsilon f(y,t,\varepsilon). \tag{18}$$

f must be a continuous, twice differentiable function of its arguments such that the limit

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

exists. A standard averaging theorem (see Chapter 8 of [15]) guarantees that, for sufficiently small ε, the solution of

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

with the same initial condition (i.e. $w(0) = y(0)$) is $O(\varepsilon)$ close to that of (18) for a time interval of length $O(\frac{1}{\varepsilon})$. What makes averaging particularly suitable for the problem at hand is, that the right hand sides of (15) and (16) are almost-periodic functions. So, we proceed to average (15) and (16). To make the procedure more transparent, we rewrite (15) in component form:

$$\dot{\chi}_i = \varepsilon\Big(\sum_{kl} H_{1kl}\,e^{i\omega_{kl}t}(\hat{\zeta}_k^*\chi_l - \chi_k^*\hat{\zeta}_l)\Big)\sum_j H_{1ij}\,e^{i\omega_{ij}t}\chi_j$$

We make two assumptions about the system:

1) The diagonal terms of the control Hamiltonian are zero, $H_{1ii} = 0, \forall i = 1,\ldots,N$. This assumption is not essential in what we are doing, but it does simplify the equations a bit.
2) All Bohr frequencies are different from each other, i.e. $\omega_{ij} \neq \omega_{kl},\ \ (i,j) \neq (k,l)$.

Since the time average of an exponential $e^{i\omega t}$ is zero, only terms with no time dependence will contribute to the averaged equations. Letting x and $\hat{z}$ be the averaged χ and $\hat{\zeta}$, the averaged state and co-state equations are:

$$\dot{x}_i = \varepsilon\sum_j |H_{1ij}|^2(x_i\hat{z}_j^* - \hat{z}_i x_j^*)x_j, \tag{19}$$

$$\dot{\hat{z}}_i = \varepsilon\sum_j |H_{1ij}|^2(x_i\hat{z}_j^* - \hat{z}_i x_j^*)\hat{z}_j. \tag{20}$$

The optimal control is then approximated up to order $O(\varepsilon)$ by

$$u = i\varepsilon \sum_{ij} H_{1ij} e^{i\omega_{ij}t}(x_j \hat{z}_i^* - \hat{z}_j x_i^*) \tag{21}$$

Note that the norm of x and $\hat{z}$ is preserved by the averaged dynamics. This is a very desirable feature, since it mimics the situation in the full dynamics.

IV. Identifying the limit $\varepsilon \to 0$ as $T \to \infty$

In equations (19) - (20), let $z = \varepsilon \hat{z}$:

$$\dot{x}_i = \sum_j |H_{1ij}|^2 (x_i z_j^* - z_i x_j^*) x_j, \tag{22}$$

$$\dot{z}_i = \sum_j |H_{1ij}|^2 (x_i z_j^* - z_i x_j^*) z_j. \tag{23}$$

Let's also write the boundary conditions (13) and (14) in terms of x and z:

$$|\psi_0' \, x(0)|^2 = 1, \quad |x_i(T)|^2 = p_i \tag{24}$$

and

$$\mathrm{Im}(\psi_0' \, z(0)) = \mathrm{Im}(x_i^*(T) z_i(T)) = 0. \tag{25}$$

A key observation is the following: If $(\, x^{(1)}(t),\, z^{(1)}(t)\,)$ is a solution of the two-point boundary value problem (22) - (25) over the time interval $[0,1]$, then

$$(\, x^{(T)}(t),\, z^{(T)}(t)\,) = (\, x^{(1)}(\frac{t}{T}),\, \frac{1}{T}\, z^{(1)}(\frac{t}{T})\,), \tag{26}$$

is a solution over the time interval $[0,T]$. It follows that,

$$\|z^{(T)}\| = \frac{\|z^{(1)}\|}{T}.$$

We can take the transfer time T large enough to make $\|z^{(T)}\|$ as small as desired. We now *postulate* the ε of the last section to be $\varepsilon = \|z^{(T)}\| = \frac{\|z^{(1)}\|}{T}$. Then $(\, x^{(T)}(t),\, z^{(T)}(t)\,)$ is, by the averaging theorem, a solution of the two-point boundary value problem (10) - (14) correct to order $\frac{1}{T}$. Although this ε and the corresponding $(\, x^{(T)}(t),\, z^{(T)}(t)\,)$ do not solve the two-point boundary value problem (10) - (14) *exactly*, they are correct up to order $\frac{1}{T}$, for T large enough. We rewrite equation (21) in the form

$$\begin{aligned} u(t) &= i \sum_{ij} H_{1ij} e^{i\omega_{ij}t}(x_j^{(T)}(t) z_i^{(T)}(t)^* - z_j^{(T)}(t) x_i^{(T)}(t)^*) \\ &= \frac{i}{T} \sum_{ij} H_{1ij} e^{i\omega_{ij}t}(x_j^{(1)}(\frac{t}{T}) z_i^{(1)}(\frac{t}{T})^* - z_j^{(1)}(\frac{t}{T}) x_i^{(1)}(\frac{t}{T})^*) \\ &= \frac{i}{T} \sum_{ij} H_{1ij} e^{i\omega_{ij}t} L_{ji}(\frac{t}{T}). \end{aligned} \tag{27}$$

Note the structure of the optimal control: It is a sum of terms of the form (exponential of a Bohr frequency) $\times$ (slow varying envelope). Moreover, the envelopes can be computed by solving the "averaged" two-point boundary value problem (22) - (25) in $[0,1]$.

We end this section with two remarks. First, the solutions we are approximating are in general local optima of the control energy. Unfortunately, except for the cases where we can solve the "averaged" (or the original) two-point boundary value problem exactly, there is usually no way to tell if a given control is *globally* energy-optimal. Second, we have not found (an approximation to) *all* solutions of the two-point boundary value problem (10) - (14). It may be the case that solutions exist such that ε is not small even for large transfer times. Such solutions, though local optima, will have too large a cost to be energy-efficient.

V. An example

Consider a three-state system with

$$H_1 = \begin{pmatrix} 0 & e^{i\alpha_{12}} & \sqrt{r}e^{i\alpha_{13}} \\ e^{-i\alpha_{12}} & 0 & e^{i\alpha_{23}} \\ \sqrt{r}e^{-i\alpha_{13}} & e^{-i\alpha_{23}} & 0 \end{pmatrix},$$

where $r < 1$. This is a system with $|(H_1)_{12}| = |(H_1)_{23}|$, where we scaled the control so that the first two matrix elements have a magnitude of 1. We take $|(H_1)_{13}| = \sqrt{r} < 1$, because we are interested in the transfer

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

and would like to see how the intermediate transitions $1 \to 2$ and $2 \to 3$ "aid" the direct transition $1 \to 3$ which has a smaller coupling. In this example, we can solve for the approximate optimal control analytically, except for a certain parameter which must be determined numerically. It turns out that the optimal control has the form

$$\begin{aligned} u(t) &= \frac{2}{T}\Big\{ a(r)\cos(\omega_{12}t + \alpha_{12} + \psi)\, \cos(\frac{\pi}{2}\frac{t}{T}) \\ &+ a(r)\sin(\omega_{23}t + \alpha_{23} + \phi - \psi)\, \sin(\frac{\pi}{2}\frac{t}{T}) \\ &+ \frac{\pi}{2(1-r)}\cos(\omega_{13}t + \phi)\Big\} \end{aligned}$$

where $\phi,\ \psi \in [0, 2\pi]$. $a(r) > 0$ has to be determined as a function of r by satisfying the boundary conditions for the state, given the form of the optimal control. Figures (1) - (3) show the approximate optimal control, the envelopes of the Bohr frequency "components" of the approximate optimal control and the averaged (dashed line) and exact (full line) evolution of the three state system under this control for $T = 20\pi$ ($r = .1$ and a is numerically found to be $a = 2.6249$). The form of the optimal control turns out to be quite intuitive: From figure (2), there is a "component" with the frequency of the transition $1 \to 3$ and constant amplitude that causes a "direct" transition from state 1 to state 3. There are also components with the frequencies of the transitions $1 \to 2$ and $2 \to 3$, such that the amplitude of the first is large initially and goes to zero at the end of the transition, while the amplitude of the second does the exact opposite. Physically, we expect a large population transfer from state 1 to state 2 which is then followed by

a large transfer from 2 to 3, and this is indeed what figure (3) demonstrates.

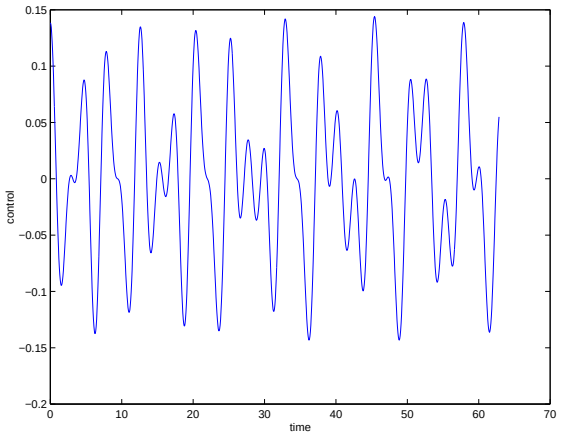

Fig. 1. Approximate optimal control for the transition $1 \rightarrow 3$ in $T = 20\pi$.

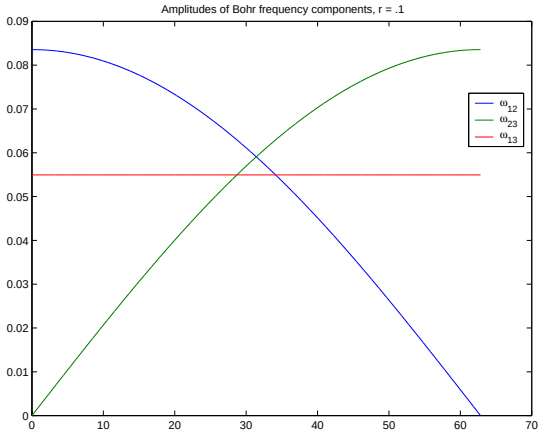

Fig. 2. Envelopes of Bohr frequency components of the approximate optimal control

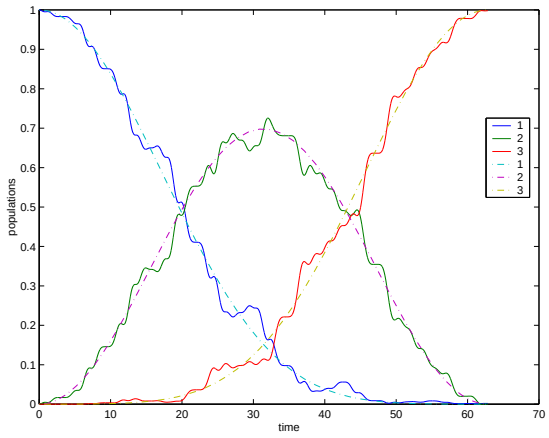

Fig. 3. Population of states vs. time. The dashed lines represent the averaged and the solid lines the exact solution.

VI. Discussion

Let's begin by summarizing our results: To find an approximate solution to the two-point boundary value problem (5) - (9) or equivalently (10) - (14), correct to order $\frac{1}{T}$ over the time horizon T, solve the two-point boundary value problem (22) - (25) over the time interval $[0, 1]$. Then, $(x^{(T)}(t), z^{(T)}(t))$ from (26) furnish the approximate solutions and (27) is the approximate optimal control. As a side remark, the size of T that makes this a good approximation is determined by the size of $\|z^{(1)}\|$. More specifically, equations (15) - (16) suggest that the quality of the approximation improves as

$$\varepsilon \|H_1\|^2 = \frac{\|z^{(1)}\|}{T} \|H_1\|^2$$

shrinks to zero.

From a theoretical standpoint, this result offers an insight into the structure of the optimal controls for incoherent population transfers. In particular, it reduces the transfer problem to a *scheduling* problem for the envelopes of the Bohr frequency sinusoids. This is both a conceptual and a computational simplification. Indeed, the original two-point boundary value problem to be solved in $[0, T]$ requires the solution of equations with dynamics "fast" on this time scale. This is because equations (5) - (6), or equivalently, (10) - (11) contain the fast dynamics of the free evolution. This is illustrated in figure (3), where the exact populations exhibit fast variations on top of their long time evolution. This makes the numerical solution of the original two-point boundary value problem very sensitive to errors, especially for large systems. On the other hand, the "averaged" two-point boundary value problem (22) - (25) does not suffer from this problem and its solution should be computationally much easier than that of (5) - (9). Furthermore, the two-point boundary value problem two-point boundary value problem (22) - (25) needs to be solved only once in order to provide (approximate) solutions to the original two-point boundary value problem for *any* transfer time T. This is in contrast to a direct numerical solution of (5) - (9) , which would have to be redone for every T. This is not to say of course that (22) - (25) constitute an easy problem to solve, especially for realistic molecular systems where the size of the system could be very big.

We would also like to comment on the relation of our work with two references, [16] and [17]. In the first work, the authors consider incoherent transfers with various convex costs depending on the moduli of the controls. They study quantum systems of the form

$$i\hbar \dot{\psi} = (H_0 + H_1(t)) \psi,$$

where each element of $H_1(t)$ is either zero or a complex control (since H_1 can have complex entries). By transforming to the "Interaction Picture", the drift term is eliminated and by redefining the controls, they end up with a driftless time-invariant control system. It then follows trivially that every control (optimal or not) is of the form, exponential of single Bohr frequency times an envelope. The main result of

that work is that the real and imaginary parts of the envelope are the same, modulo a phase, for optimal transfers. The point is then that one can reduce the number of controls one is optimizing over. In our case, the controls enter in a more general fashion, and, in general, there are not enough controls so that, after eliminating the drift, we end up with a driftless time-invariant system. Rather, the form of the optimal control is a consequence of the large transfer time limit.

In the second work, the authors consider (3) *linearized* around $(\psi^{(0)}(t), u^{(0)}(t)) = (e^{-iE_i t}e_i, 0)$, an equilibrium point of the free dynamics. They consider optimal coherent state transfers for this linearized system with a quadratic cost and find that the resulting optimal control is a mix of sinusoidals with Bohr frequencies $\omega_{ji},\ j \neq i$. The approximation of (3) by a linearized model is valid for transfer times of order 1 and final states ε close to the initial one, where ε is the expansion parameter. Our result concerns the exact model, arbitrary initial and final populations and holds for times of order $\frac{1}{\varepsilon} \sim T$. Again, the special form of the resulting optimal control is a consequence of the large transfer time limit.

Finally, we would like to mention some possible extensions of this work. An obvious direction is the search for a computationally efficient procedure to solve the two-point boundary value problem (22) - (25), especially for large systems. Questions of robustness to parameter uncertainty and noise in the control implementation have to be addressed as well.

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