

ANALYSIS OF LYAPUNOV CONTROL FOR HAMILTONIAN QUANTUM SYSTEMS

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Abstract

We present detailed analysis of the convergence properties and effectiveness of Lyapunov control design for bilinear Hamiltonian quantum systems based on the application of LaSalle's invariance principle and stability analysis from dynamical systems and control theory. For a certain class of Hamiltonians, strong convergence results can be obtained for both pure and mixed state systems. The control Hamiltonians for realistic physical systems, however, generally do not fall in this class. It is shown that the effectiveness of Lyapunov control design in this case is significantly diminished.

Key words

Quantum systems, dynamical systems, control design, Lyapunov functions

1 Introduction

Control of quantum phenomena is becoming increasingly important in many divergent areas of research including quantum computation, quantum chemistry, nano-scale materials, and Bose-Einstein condensates. Accordingly, quantum control theory has developed greatly in both depth and breath in recent years, and many results about controllability and control methods have been obtained. Broadly speaking, quantum control approaches fall into two categories: open-loop Hamiltonian (and sometimes reservoir) engineering using a variety of techniques including optimal control [Shi, Woody and Rabitz, 1998; Maday and Turinici, 2003; Schirmer, Girardeau and Leahy, 2000] and geometric designs [Jurdjevic, 1997; Jurdjevic and Sussmann, 1972; Koch and Lowenthal, 1975; D'alessandro, 2000; Schirmer, 2002], and closed-loop quantum state reduction and stabilization using feedback from weak measurements [Wiseman and Milburn, 1993; Wiseman, 1994].

Lyapunov functions have played a significant role in control design. Originally used in feedback control to analyze the stability of the controlled system,

they have formed the basis for new control designs, and several recent papers have discussed the application of Lyapunov control designs to quantum systems [Vettori, 2002; Ferrante, Pavon and Raccanelli, 2002; Grivopoulus and Bamieh, 2003; Mirrahimi and Rouchon, 2004a; Mirrahimi and Rouchon, 2004b; Mirrahimi and Turinici, 2005; Altafini, 2007a; Altafini, 2007b]. Although the basic mathematical formalism is well established, using either the Schrodinger equation for pure state wavefunctions [Vettori, 2002; Ferrante, Pavon and Raccanelli, 2002; Grivopoulus and Bamieh, 2003], or the Liouville-von Neumann equation [von-Neumann, 1955] for density operators [Altafini, 2007a; Altafini, 2007b], many questions remain. For example, some sufficient conditions for the method to be effective, i.e., guarantee convergence of the system state converges to the target state, have been obtained [Mirrahimi and Rouchon, 2004a; Mirrahimi and Rouchon, 2004b; Mirrahimi and Turinici, 2005; Altafini, 2007a; Altafini, 2007b], but are they also necessary? What are explicit requirements on the Hamiltonian and the target state such that the control is effective?

Moreover, there are outstanding technical issues. The invariance principle [LaSalle and Lefschetz, 1961] should be applied to autonomous systems, and its application for time-dependent target states needs careful justification. Moreover, the trajectory of time-dependent target state under free evolution is generally not periodic, as previously asserted in the literature [Altafini, 2007b]. We address these issues and present a detailed analysis of the relationship between the effectiveness of Lyapunov control design and the parameters in the control problem, the Hamiltonian and the target state. In section II, we establish the mathematical model for the quantum system controlled by Lyapunov method, and apply LaSalle invariance principle to analyze the convergence condition. In section III and IV, we will use the results in section II to discuss the Lyapunov control of pseudo-pure state and generic state, under an ideal condition of the Hamiltonian; in section V, we shall relax this condition and see how the effectiveness of Lyapunov control will change.

2 Control System and Invariance Principle

2.1 Controlled dynamics and Lyapunov function

A controlled quantum system can be modelled in different ways, either as a closed system evolving unitarily under certain Hamiltonian, or as an open system interacting with a heat bath. In this paper, we restrict our discussion to an n -level bilinear Hamiltonian dynamical system satisfying the Liouville-von Neumann equation: (assuming $\hbar = 1$):

$$\dot{\rho}(t) = -i[H_0 + f(t)H_1, \rho(t)], \quad (1)$$

where ρ is a positive trace-one operator, representing the system state, H_0 is the free evolution Hamiltonian and H_1 is the controlled Hamiltonian, both of which are constant. In the special case when the system is in a pure state $|\psi\rangle$, we have $\rho = |\psi\rangle\langle\psi|$, and the dynamical system can be represented as:

$$\dot{\psi}(t) = -i(H_0 + f(t)H_1)\psi(t) \quad (2)$$

although we will use the density operator formulation throughout this paper. The general control task we consider can be formulated as, given a target state ρ_d , we wish to apply a certain control field $f(t)$ to the system that modifies its dynamics such that $\rho(t) \rightarrow \rho_d$ as $t \rightarrow +\infty$. Since the free Hamiltonian H_0 can generally not be turned off, it is natural to assume ρ_d to be time-dependent, satisfying:

$$\dot{\rho}_d(t) = -i[H_0, \rho_d(t)]. \quad (3)$$

Since the evolution of both $\rho(t)$ and $\rho_d(t)$ is unitary in our case, we require $\rho(0)$ to be unitarily equivalent to $\rho_d(0)$. Hence, the state space \mathcal{M} is the set of all density operators ρ such that ρ and $\rho_d(0)$ are unitarily equivalent. This is a compact manifold, called a flag manifold, whose dimension depends on the number of distinct eigenvalues of ρ_d . We say ρ_d is pseudo-pure if its spectrum has only two distinct values, one occurring with multiplicity one and the other with multiplicity $n - 1$, and ρ_d is generic if it has n non-degenerate eigenvalues. We have $2n - 2 \leq \dim(\mathcal{M}) \leq n^2 - n$ with $\dim(\mathcal{M}) = n^2 - n$ for generic ρ_d and $\dim(\mathcal{M}) = 2n - 2$ for pure or pseudo-pure states.

Define a function V on $\mathcal{M} \times \mathcal{M}$:

$$\begin{aligned} V(\rho, \rho_d) &= \frac{1}{2} \text{Tr}((\rho - \rho_d)^2) \\ &= \text{Tr}(\rho_d^2) - \text{Tr}(\rho_d \rho). \end{aligned} \quad (4)$$

We have $V \geq 0$ with equality only if $\rho = \rho_d$. Taking derivative of V along any solution $(\rho(t), \rho_d(t))$, and

substituting (1) and (3), we derive:

$$\begin{aligned} \dot{V} &= -\text{Tr}(\dot{\rho}_d \rho) - \text{Tr}(\rho_d \dot{\rho}) \\ &= -\text{Tr}([-iH_0, \rho_d] \rho) - \text{Tr}(\rho_d [-iH_0, \rho]) \\ &\quad - f(t) \text{Tr}(\rho_d [-iH_1, \rho]) \\ &= -f(t) \text{Tr}(\rho_d [-iH_1, \rho]). \end{aligned}$$

If we choose $f(t) = \kappa \text{Tr}(\rho_d [-iH_1, \rho])$, $\kappa > 0$, then $\dot{V}(\rho(t), \rho_d(t)) \leq 0$. Hence, V is a Lyapunov function for the following autonomous dynamical system with respect to $(\rho(t), \rho_d(t))$:

$$\begin{aligned} \dot{\rho}(t) &= -i[H_0 + f(\rho, \rho_d)H_1, \rho(t)] \\ \dot{\rho}_d(t) &= -i[H_0, \rho_d(t)] \\ f(\rho, \rho_d) &= \kappa \text{Tr}([-iH_1, \rho] \rho_d) \end{aligned} \quad (5)$$

2.2 LaSalle invariance principle and invariant set

To complete the control task, we require $\rho(t) \rightarrow \rho_d(t)$ as $t \rightarrow +\infty$, which is equivalent to $V(\rho(t), \rho_d(t)) \rightarrow 0$. A key result for the convergence analysis is LaSalle's invariance principle [LaSalle and Lefschetz, 1961]:

Theorem 2.1. *Let $V(x)$ be a Lyapunov function on the phase space $\Omega = \{x\}$ of an autonomous dynamical system $\dot{x} = f(x)$, satisfying $V(x) > 0$ for all $x \neq x_0$ and $\dot{V}(x) \leq 0$. Let $\mathcal{O}(\bar{x}(t))$ be the orbit of $\bar{x}(t)$ in Ω . Then the invariant set $E = \{\mathcal{O}(x(t)) | \dot{V}(\bar{x}(t)) = 0\}$, contains the positive limiting sets of all bounded solutions, i.e., any bounded solution converges to E as $t \rightarrow +\infty$.*

For our quantum dynamical system (5), since the state space \mathcal{M} is compact, any solution $(\rho(t), \rho_d(t))$ is bounded. Applying LaSalle Invariance Principle we obtain [Wang and Schirmer, 2008]:

Theorem 2.2. *The state $(\rho(t), \rho_d(t))$ of the autonomous dynamical system (5) converges to the invariant set $E = \{(\rho_1, \rho_2) \in \mathcal{M} \times \mathcal{M} | \dot{V}(\rho(t), \rho_d(t)) = 0, (\rho(0), \rho_d(0)) = (\rho_1, \rho_2)\}$.*

Therefore, the next step is to determine the invariant set E , for the dynamical system (5). Notice that in LaSalle invariance principle, E contains the positive limiting points of all bounded solutions for any $\rho_d(t)$. Hence, in the following, we always restrict the calculation of E to the points (ρ_1, ρ_2) such that ρ_2 is the positive limiting point of the given $\rho_d(t)$. For our dynamical system, the invariant set $E = \{V(\rho(t), \rho_d(t)) = 0\}$ is equivalent to $f(t) = 0$, for any t :

$$\begin{aligned} 0 &= f = \text{Tr}([-iH_1, \rho] \rho_d) \\ 0 &= \dot{f} = \text{Tr}([-iH_1, \rho] \dot{\rho}_d) + \text{Tr}([-iH_1, \dot{\rho}] \rho_d) \\ &= -\text{Tr}([[-iH_0, -iH_1], \rho] \rho_d) \\ &\dots \\ 0 &= \frac{d^\ell f}{dt^\ell} = (-1)^\ell \text{Tr}([Ad_{-iH_0}^\ell(-iH_1), \rho] \rho_d), \end{aligned}$$

where $Ad_{-iH_0}^\ell(-iH_1)$ represents ℓ -fold commutator adjoint action of $-iH_0$ on $-iH_1$. Hence, $\text{Tr}([A, B]C) = -\text{Tr}([C, B]A) = -\text{Tr}([A, C]B)$ gives a necessary condition for the invariant set E :

$$\text{Tr}([\rho, \rho_d] Ad_{-iH_0}^m(-iH_1)) = 0, \quad (6)$$

where $Ad_{-iH_0}^0(-iH_1) = -iH_1$ and m is any non-negative integer. Hence the invariant E depends on both the Hamiltonian and the target state. Without loss of generality, we assume H_0 and H_1 to be trace-zero. Since H_0 is hermitian and therefore diagonalizable, we may assume $H_0 = \text{diag}(a_1, \dots, a_n)$, with diagonal elements arranged in decreasing order, where the diagonal elements physically represent the energy levels of the system. We shall assume H_1 to be off-diagonal in this basis with off-diagonal elements $b_{k\ell}$ representing the couplings between the energy levels k and ℓ . Also let $\omega_{k\ell} = a_\ell - a_k$ be the transition frequency between the energy levels k and ℓ . With these assumptions, we can prove the following useful theorem [Wang and Schirmer, 2008]:

Theorem 2.3. *If (1) H_0 strongly regular, i.e., $\omega_{k\ell} \neq \omega_{pq}$ unless $(k, \ell) = (p, q)$, and (2) H_1 fully connected, i.e., $b_{k\ell} \neq 0$ except for $k = \ell$, then (ρ_1, ρ_2) belongs to the invariant set E if and only if $[\rho_1, \rho_2]$ is diagonal.*

We note that these conditions on the Hamiltonian are very strong and rarely satisfied for real physical systems. However, this is the ideal case for Lyapunov control design, and it is useful to begin by analyzing the effectiveness of the method in this ideal case before relaxing the requirements.

2.3 Real representation for quantum systems

In order to apply stability analysis to our complex quantum dynamical system, we require a real representation for both the Hamiltonian and the density operator. Let $\mathcal{B}_{\mathbb{R}}(\mathcal{H})$ be the real vector space of all $n \times n$ Hermitian matrices on the Hilbert space \mathcal{H} . For any $H_1, H_2 \in \mathcal{B}_{\mathbb{R}}(\mathcal{H})$, we can define an inner product $\langle H_1 | H_2 \rangle = \text{Tr}(H_1 H_2)$, and an associated orthonormal basis $\{\lambda_k, \lambda_{k\ell}, \bar{\lambda}_{k\ell}\}$, where

$$\lambda_0 = \frac{1}{\sqrt{n}}(\hat{e}_{11} + \hat{e}_{22} + \dots + \hat{e}_{n,n}) \quad (7a)$$

$$\lambda_k = \frac{1}{\sqrt{k(k+1)}}(\hat{e}_{11} + \dots + \hat{e}_{kk} - k\hat{e}_{k+1,k+1}) \quad (7b)$$

$$\lambda_{k\ell} = \frac{1}{\sqrt{2}}(\hat{e}_{k\ell} + \hat{e}_{\ell k}) \quad (7c)$$

$$\bar{\lambda}_{k\ell} = \frac{i}{\sqrt{2}}(-\hat{e}_{k\ell} + \hat{e}_{\ell k}), \quad (7d)$$

$\hat{e}_{k\ell}$ being the elementary matrix with 1 in the (k, ℓ) position and 0 elsewhere, and $1 \leq k < \ell \leq n$. In this basis, any hermitian matrix H can be represented as an n^2 -dimensional real vector. For density operators ρ with $\text{Tr}(\rho) = 1$ the coefficient of λ_0 is constant and thus can be dropped. Let $\vec{s}(t)$ and $\vec{s}_d(t)$ be the vectors in \mathbb{R}^{n^2-1} representing $\rho(t)$ and $\rho_d(t)$. The adjoint

action $\text{Ad}_{iH}(\rho) = [iH, \rho]$ in this basis is given by an anti-symmetric matrix A acting on $\vec{s}(t)$. Therefore, the quantum dynamical system (5) can be equivalently represented as:

$$\begin{aligned} \dot{\vec{s}}(t) &= (A_0 + A_1)\vec{s}(t) \\ \dot{\vec{s}}_d(t) &= A_0\vec{s}_d(t) \\ f(t) &= \vec{s}_d^T A_1 \vec{s} \end{aligned} \quad (8)$$

where $A_0 = \text{Ad}_{-iH_0}$ and $A_1 = \text{Ad}_{-iH_1}$. This real representation is generally known as the Bloch vector form especially for $n = 2$.

3 Convergence Analysis for Ideal Systems

In this section we consider ideal systems, i.e., systems with H_0 strongly regular and H_1 fully connected. If the system and hence the Hamiltonian are fixed, the invariant set E depends on the target state ρ_d only. In general, the convergence analysis depends on the spectrum of the target state ρ_d , in particular the multiplicities of its eigenvalues. As we cannot give a complete discussion of all possible cases here, we will focus on two cases of crucial importance, (a) when ρ_d is a generic mixed state with n non-degenerate eigenvalues, and (b) when ρ_d is a pure or pseudo-pure state with only two eigenvalues with multiplicities 1 and $n - 1$, respectively, starting with the two-level case.

3.1 Pseudo-pure states

We start with the special case of a two-level system, for which the embedding of density operators into \mathbb{R}^{n^2-1} gives rise to a *homeomorphism* between density operators and points inside a closed ball in \mathbb{R}^3 , with pure states forming the surface of the ball, and the completely mixed state $(0, 0, 0)$ its centre. In this case there is no distinction between pseudo-pure and generic states, all states except the completely mixed state $\rho_0 = \text{diag}(\frac{1}{2}, \frac{1}{2})$ being both generic and pseudo-pure. Furthermore, the requirements of strong regularity of H_0 and full connectedness of H_1 are always satisfied for a two-level systems, except for the trivial cases of systems with a single degenerate state or no coupling to the control field. Excluding these trivial cases, Theorem 2.3 implies that all $(\rho_1, \rho_2) \in E$ satisfy $[\rho_1, \rho_2]$ diagonal, and one can show that there are three types of points in the invariant set [Wang and Schirmer, 2008]

- (a) $\text{Tr}(\rho_1 \rho_2) = 1$, i.e., $\rho_1 = \rho_2$;
- (b) $\text{Tr}(\rho_1 \rho_2) = 0$;
- (c) $\text{Tr}[\lambda_1 \rho_1] = \text{Tr}[\lambda_1 \rho_2] = 0$.

Another special feature of the $n = 2$ case is that for any choice of H_0 , the trajectory of $\rho_d(t)$ forms a periodic orbit $\mathcal{O}(\rho_d(0))$, which is a compact set, so any positive limiting point (ρ_1, ρ_2) of $(\rho(t), \rho_d(t))$ must satisfy $\rho_2 \in \mathcal{O}(\rho_d(0))$. Therefore, if $\rho_d(0)$ has nonzero λ_1 component, i.e., $\text{Tr}(\rho_d(0)\lambda_1) \neq 0$ then E can only

contain the cases (a) and (b), corresponding to the values of Lyapunov function $V = V_{\max}$ and $V = 0$. Hence, for any $\rho(t)$ with $\text{Tr}(\rho(0)\rho_d(0)) \neq 0$, we have $V(\rho(0), \rho_d(0)) < V_{\max}$ and LaSalle's invariance principle guarantees that $V(\rho(t), \rho_d(t)) \rightarrow 0$ and $\rho(t) \rightarrow \rho_d(t)$ as $t \rightarrow +\infty$. Lyapunov control in this case is an effective strategy.

If $\rho_d(0)$ has zero λ_1 component, however, then the invariant set E contains all points $(\rho_1\rho_2)$ satisfying (c), the value of the Lyapunov function V on E spans the entire interval $[0, V_{\max}]$, and we cannot conclude that $\rho(t) \rightarrow \rho_d(t)$. Indeed simulations suggest that V can tend to any value in $[0, V_{\max}]$ in this case. We can still conclude that $\rho(t)$ converges to the set $\mathcal{O}(\rho_d(t))$ corresponding to the orbit of $\rho_d(t)$ but this is a substantially weaker notion of convergence as there are infinitely many distinct states whose orbits under free evolution coincide.

If we take the Bloch vector to be $\vec{s} = (x, y, z)$ with $x = \text{Tr}(\rho\lambda_{12})$, $y = \text{Tr}(\rho\bar{\lambda}_{12})$ and $z = \text{Tr}(\rho\lambda_1)$, as usual for $n = 2$, then case (a) corresponds to $\vec{s}_1 = \vec{s}_2$, case (b) corresponds to \vec{s}_1 being antipodal to \vec{s}_2 , $\vec{s}_1 = -\vec{s}_2$, and (c) corresponds to the target state lying on the equator of the sphere. If $\vec{s}_d(0)$ is not on the equator, all solutions $\vec{s}(t)$ with $\vec{s}(0) \neq -\vec{s}_d(0)$ converge to $\vec{s}_d(t)$. If $\vec{s}_d(0)$ is on the equator, any solution $\vec{s}(t)$ will converge to the equator but $\vec{s}(t) \not\rightarrow \vec{s}_d(t)$ in general. The picture is the same for all equivalence classes of states, except the completely mixed state, the only difference being that pure states lie on the surface of the ball, while pseudo-pure states with the same spectrum lie on concentric spherical shells of in the interior.

For $n > 2$ pseudo-pure states are exceptional or non-generic and the mapping from density operators into \mathbb{R}^{n^2-1} provided by the Bloch vector is only an embedding, not a homeomorphism. Furthermore, the conditions on the Hamiltonian of strong regularity and complete connectedness are less trivial in this case as there are many systems that are connected and regular and controllable, but not strongly regular or fully connected. However, assuming such ideal Hamiltonians we can still prove [Wang and Schirmer, 2008]:

Theorem 3.1. *Given a pseudo-pure state target state $\rho_d(t)$ with spectrum $\{w, u\}$ and ideal Hamiltonians as defined above, Lyapunov control is effective, i.e., any solution $\rho(t)$ with $V(\rho(0), \rho_d(0)) < V_{\max}$ will converge to $\rho_d(t)$ as $t \rightarrow +\infty$, except when ρ_d has a single pair of non-zero off-diagonal entries of the form $r_{k\ell}(t) = \frac{1}{2}(w - u)e^{i\alpha}$ and $r_{kk} = r_{\ell\ell} = \frac{1}{2}(w + u)$. In the latter case any solution $\rho(t)$ will converge to the orbit of $\rho_d(t)$ but in general $\rho(t) \not\rightarrow \rho_d(t)$ as $t \rightarrow +\infty$ and $V(\rho, \rho_d)$ can take any limiting value between 0 and V_{\max} .*

This theorem essentially asserts that if H_0 is strongly regular, H_1 is fully connected and ρ_d is a pseudo-pure state whose dynamics is not confined to a periodic orbit in a two-dimensional subspace, then any solution $\rho(t)$ with $\text{Tr}(\rho(0)\rho_d(0)) \neq 0$ will converge to $\rho_d(t)$ as

$t \rightarrow +\infty$. Thus for $n > 2$ the special case where $\rho(t)$ converges to the orbit $\mathcal{O}(\rho_d(t))$ but not $\rho_d(t)$, corresponding to case (c) above, is precluded, except when the target state is such that its orbit is a periodic orbit (circle) in a 2D subspace. Given any other pseudo-pure state $\rho_d(t)$, all initial states $\rho(0)$ that are not part of the critical manifold of states for which $\text{Tr}(\rho(0)\rho_d(0)) = 0$ and V assumes its maximum, will converge to the target state $\rho_d(t)$ for $t \rightarrow \infty$.

3.2 Generic states for n -level systems

For $n > 2$ pseudo-pure states are a very small subset of the state space. Most states ρ are generic with n non-degenerate eigenvalues. We distinguish two cases here: stationary target states ρ_d , which are diagonal (in the eigenbasis of H_0), and non-stationary target states. When ρ_d is stationary, the dynamical system (5) can be reduced to

$$\begin{aligned} \dot{\rho}(t) &= -i[H_0 + f(\rho)H_1, \rho(t)] \\ f(\rho) &= \text{Tr}([-iH_1, \rho(t)]\rho_d) \end{aligned} \quad (9)$$

and the invariant set (for an ideal system) reduces accordingly to $E = \{\rho_0 | \dot{V}_{\rho_d}(\rho(t)) = 0, \rho(0) = \rho_0\}$, which can be shown to be equivalent to the set of all ρ_0 with $[\rho_0, \rho_d]$ diagonal. Furthermore, since ρ_d is stationary and thus diagonal, the latter condition can further be reduced to $[\rho_0, \rho_d] = 0$ by virtue of the following:

Lemma 3.1. *If A is diagonal with non-degenerate eigenvalues and $[A, B]$ is diagonal, then B is also diagonal and $[A, B] = 0$.*

The proof follows trivially from the fact that for $A = \text{diag}(a_1, \dots, a_n)$ and $B = (b_{mn})$, the (m, n) component of $[A, B]$ is $b_{mn}(a_m - a_n)$. If $[A, B]$ is diagonal and $a_m \neq a_n$ then we must have $b_{mn} = 0$ for $m \neq n$. This leads to the following [Wang and Schirmer, 2008]:

Theorem 3.2. *If ρ_d is a generic stationary target state then the invariant set E contains exactly the $n!$ critical points of the Lyapunov function $V(\rho) = \text{Tr}(\rho_d^2) - \text{Tr}(\rho\rho_d)$, i.e., the stationary states $\rho_d^{(k)}$, $k = 1, \dots, n!$, which commute with ρ_d and have the same spectrum.*

As ρ_d is stationary and therefore diagonal, it follows that all $\rho_d^{(k)}$ are also diagonal, and their diagonal elements are a permutation of those of ρ_d . Since $\text{Tr}(\rho_d^2)$ is constant for a Hamiltonian system, the critical points of $V(\rho)$ coincide with the critical points of $J(\rho) = \text{Tr}(\rho_d\rho)$, which can be regarded as the expectation value of the observable $A = \rho_d$. We can immediately see that V assumes its global minimum when the expectation value of $\text{Tr}(\rho_d\rho)$ assumes its maximum, i.e., for $\rho = \rho_d$, and its maximum when $J(\rho)$ assumes its minimum. Assuming $\rho_d^{(0)} = \rho_d = \text{diag}(w_1, \dots, w_n)$ and $\rho_d^{(n!)} = \text{diag}(w_{\tau(1)}, \dots, w_{\tau(n)})$, where τ is the

permutation of $\{1, \dots, n\}$ that corresponds to a complete inversion, i.e., $\tau(k) = n + 1 - k$, we have [Girardeau, Schirmer, Leahy and Koch, 1998]:

$$J(\rho_d^{(n!)}) \leq J(\rho) \leq J(\rho_d^{(1)}), \quad (10)$$

i.e., $\rho_d^{(0)}$ and $\rho_d^{(n!)}$ correspond to the global extrema of V with $V = 0$ and $V = V_{\max} = \sum_k w_k^2$, respectively.

It is furthermore easy to show that for a given generic stationary state ρ_d the critical points of the Lyapunov function $V(\rho)$ are hyperbolic. However, since the dynamical system defined by our Lyapunov control is not the gradient flow of $V(\rho)$, asymptotic stability of these fixed points can not be derived directly from the associated index number of the Morse function V . Nonetheless, further analysis of the linearization of the dynamics near the critical points shows that [Wang and Schirmer, 2008]:

Theorem 3.3. *For a generic stationary target state ρ_d all the critical points of the dynamical system (9) are hyperbolic. ρ_d is the only sink, all other critical points are saddles, except the global maximum, which is a source.*

Since the critical points of the dynamical system (5) for a generic stationary state ρ_d are hyperbolic and they are also hyperbolic critical points of the function $V(\rho) = V(\rho, \rho_d)$, the dimension of the stable manifold at a critical point must be the same as the index number of the critical point of the function V . In particular, since all critical points except the global minimum and maximum are saddle points, they are not repulsive, and therefore there are solutions $\rho(t)$ outside E that converge to these saddles, resulting in the failure of the Lyapunov control method. However, as the dimensions of the stable manifolds at these points are smaller than $\dim(\mathcal{M})$, almost all solutions will still converge to the global minimum $\rho_d^{(1)} = \rho_d$, and thus the Lyapunov method is still (mostly) effective.

When the target state ρ_d is not stationary, the situation is somewhat more complicated as the invariant set E may contain points with nonzero diagonal commutators.

Example 3.1. *Consider ρ_1 and $\rho_d(0) = \rho_2$ with*

$$\rho_1 = \begin{pmatrix} \frac{1}{12} & -\frac{1}{12} & -\frac{1}{12} \\ -\frac{1}{12} & \frac{11}{24} & \frac{1}{8} \\ -\frac{1}{12} & \frac{1}{8} & \frac{11}{24} \end{pmatrix}, \quad \rho_2 = \begin{pmatrix} \frac{1}{3} & -\frac{i}{12} & \frac{i}{12} \\ \frac{i}{12} & \frac{1}{3} & -\frac{i}{4} \\ -\frac{i}{12} & \frac{i}{4} & \frac{1}{3} \end{pmatrix}.$$

ρ_1 and ρ_2 are isospectral and we have

$$[\rho_1, \rho_2] = \begin{pmatrix} 0 & 0 & 0 \\ 0 & \frac{11}{144}i & 0 \\ 0 & 0 & -\frac{11}{144}i \end{pmatrix}$$

i.e., $(\rho_1, \rho_2) \in E$ but $[\rho_1, \rho_2] \neq 0$.

Simulations suggest that $\rho(t)$ does not converge to $\rho_d(t)$ or $\mathcal{O}(\rho_d(t))$ in this case and thus Lyapunov control fails. It is difficult to give a rigorous proof of this observation, however, as we lack a constructive method to ascertain asymptotic stability near a non-stationary solution. In the special case where $\rho_d(t)$ is periodic there are tools such as Poincaré maps but it is difficult to write down an explicit form of the Poincaré map for general periodic orbits [Perko, 2000]. Moreover, as observed earlier, for $n > 2$ the orbits of non-stationary target states $\rho_d(t)$ under H_0 are periodic only in some exceptional cases.

Fortunately, however, $E = \{[\rho_1, \rho_2] = 0\}$ still holds for a very large class of generic target states $\rho_d(t)$, and in these cases Lyapunov control tends to be effective. Setting $[\rho_1, \rho_2] = -\text{Ad}_{\rho_2}(\rho_1)$, where Ad_{ρ_2} is a linear map from the Hermitian or anti-Hermitian matrices into $\mathfrak{su}(n)$, let $A(\vec{s}_2)$ be the real $(n^2 - 1) \times (n^2 - 1)$ matrix corresponding to the Stokes representation of Ad_{ρ_2} . Recall $\mathfrak{su}(n) = \mathcal{T} \oplus \mathcal{C}$ and $\mathbb{R}^{n^2-1} = S_{\mathcal{T}} \oplus S_{\mathcal{C}}$, where $S_{\mathcal{C}}$ and $S_{\mathcal{T}}$ are the real subspaces corresponding to the Cartan and non-Cartan subspaces, \mathcal{C} and \mathcal{T} , respectively. Let $\hat{A}(\vec{s}_2)$ be the first $n^2 - n$ rows of $A(\vec{s}_2)$ (whose image is $S_{\mathcal{T}}$). Then we can show [Wang and Schirmer, 2008]:

Theorem 3.4. *The invariant set E for a generic $\rho_d(t)$ contains points with nonzero commutator only if either ρ_d has some equal diagonal elements or $\det(\hat{A}_1) = 0$. Therefore, the set of $\rho_d(0)$ such that E contains points with nonzero commutator has measure zero with respect to the state space \mathcal{M} .*

Hence, if we choose a generic target state $\rho_d(0)$ randomly, with probability one, it will be such that $E = \{[\rho_1, \rho_2] = 0\}$. Choosing an orthonormal basis such that ρ_2 is diagonal, it thus follows that ρ_1 must be diagonal in this basis, and its diagonal elements a permutation of the eigenvalues of ρ_2 . Thus for a given target state $\rho_2 = \rho_d$, there are again $n!$ critical points $(\rho_d^{(k)}(t), \rho_d(t))$. Moreover, for any $(\rho_1, \rho_2) \in E$ with $\rho_2 = \rho_d$ and $\rho_1 = \rho_2^{(k)}$ for some k , there exists a subsequence $\{t_n\}$ such that $(\rho(t_n), \rho_d(t_n)) \rightarrow (\rho_1, \rho_2)$. In particular, $\rho(t_n) \rightarrow \rho_1$, $\rho_d(t_n) \rightarrow \rho_2$, and hence, $\rho_d^{(k)}(t_n) \rightarrow \rho_2^{(k)} = \rho_1$. Therefore, we have $\rho(t_n) \rightarrow \rho_d^{(k)}(t_n)$. If $(\rho_1, \rho_2) \in E$ is a different positive limiting point of $(\rho(t), \rho_d(t))$, since $V(\rho(t), \rho_d(t))$ decreases along the trajectory and the $n!$ critical points are isolated, we must have $\rho_1 = \rho_2^{(k)}$ for the same k as for (ρ_1, ρ_2) . Therefore, $\rho(t_n) \rightarrow \rho_d^{(k)}(t_n)$ holds for any positive limiting point (ρ_1, ρ_2) and the corresponding subsequence $\{t_n\}$. Hence $\rho(t) \rightarrow \rho_d^{(k)}(t)$ as $t \rightarrow +\infty$, and some further analysis shows [Wang and Schirmer, 2008]:

Theorem 3.5. *If $\rho_d(t)$ is a generic state with invariant set $E = \{[\rho_1, \rho_2] = 0\}$ then any solution $\rho(t)$ converges to one of the $n!$ critical points $\rho_d^{(k)}(t)$, and all*

solutions except $\rho_d^{(1)}(t) = \rho_d(t)$, which is stable, are unstable.

Thus we have a similar result as for generic stationary ρ_d . The difference is that for the stationary ρ_d , we can present a quantitative result about the dimensions of the stable manifolds and hence the measure of solutions that will converge to these points. For the non-stationary target case we can not establish the analogous result. However, simulations suggest that almost all solutions will converge to $\rho_d(t)$, and we can further prove a weaker result [Wang and Schirmer, 2008]:

Proposition 3.1. *For any of the unstable solutions, $\rho_d^{(k)}$, $k = 2, \dots, n! - 1$, we can find solutions $\rho(t)$, with $V(\rho(0), \rho_d(0)) > V(\rho_d^{(k)}(0), \rho_d(0))$, that still converge to $\rho_d(t)$.*

4 Effectiveness of Method for Non-ideal Systems

The previous analysis relied on strong assumptions about the system, assuming H_0 strongly regular and H_1 fully connected. We shall now relax these requirements to see how the invariant set E and the effectiveness of the Lyapunov control method change. Without loss of generality, we present the analysis for a three-level system, noting that the generalization to n -level systems is straightforward.

First suppose H_0 is strongly regular, as for a Morse oscillator, for example, but some of the off-diagonal elements of H_1 are zero, corresponding to transitions with zero transition probability. This is the case for many physical systems, where often only transitions between adjacent energy levels are permitted, and we rarely have non-vanishing transition probabilities for all possible transitions. For concreteness, assume $n = 3$ and

$$H_0 = \begin{pmatrix} a_1 & 0 & 0 \\ 0 & a_2 & 0 \\ 0 & 0 & a_3 \end{pmatrix}, \quad H_1 = \begin{pmatrix} 0 & b_1 & 0 \\ b_1^* & 0 & b_2 \\ 0 & b_2^* & 0 \end{pmatrix},$$

where $a_1 > a_2 > a_3$. In this case, we can prove [Wang and Schirmer, 2008] that any point $(\rho_1, \rho_2) \in E$ must satisfy:

$$[\rho_1, \rho_2] = \begin{pmatrix} \alpha_{11} & 0 & e^{-i\omega_{13}t}\alpha_{13} \\ 0 & \alpha_{22} & 0 \\ e^{i\omega_{13}t}\alpha_{13}^* & 0 & \alpha_{33} \end{pmatrix}.$$

Hence, for a stationary and generic target state ρ_d the points ρ_1 in E must have the form

$$\rho_1 = \begin{pmatrix} \beta_{11} & 0 & \beta_{13} \\ 0 & \beta_{22} & 0 \\ \beta_{13}^* & 0 & \beta_{33} \end{pmatrix}.$$

We can prove that near the stationary point ρ_d , the invariant set E forms a centre manifold with ρ_d as

a centre [Wang and Schirmer, 2008]. Therefore, the Hartman-Grobman theorem from the centre manifold theory, proved by Carr [Carr, 1981], implies that almost all solutions near $\rho_d(t)$ converge to the (periodic) solutions on the centre manifold instead of the centre ρ_d . Lyapunov method in this case is ineffective.

Second, we consider the case of H_1 fully connected but H_0 is not strongly regular. For example, consider

$$H_0 = \begin{pmatrix} a_1 & 0 & 0 \\ 0 & a_2 & 0 \\ 0 & 0 & a_3 \end{pmatrix}, \quad H_1 = \begin{pmatrix} 0 & b_1 & b_3 \\ b_1^* & 0 & b_2 \\ b_3^* & b_2^* & 0 \end{pmatrix}$$

with $\omega_{12} = \omega_{23}$, where $\omega_{mn} = a_m - a_n$. In this case we can also prove that the invariant set E forms a centre manifold near ρ_d with ρ_d as a centre, and thus that almost all solutions near $\rho_d(t)$ will converge to the periodic solutions on the centre manifold other than the centre ρ_d , and hence that the Lyapunov method is still ineffective.

In summary, when either of the stringent conditions on the Hamiltonians are relaxed even slightly, the invariant set E becomes much larger, and in marked contrast to the ideal system case, and the Lyapunov method fails for almost all cases.

5 Conclusion and remarks

We have presented a detailed analysis of the Lyapunov control method for bilinear quantum control systems based on the application of the LaSalle invariance principle. For the case of non-stationary target states, this required considering the dynamics on an augmented state space on which the total system is autonomous. Characterization and analysis of the invariant set of this dynamical system allowed us to establish a quite clear picture of the effectiveness of the Lyapunov method depending on the properties of the system and the target state. In particular, our analysis suggests that the method is generally only effective under very stringent assumptions on the Hamiltonian, and even in this case our analysis suggests a rather more complicated picture than previously presented in the literature (see e.g. [Altafini, 2007a; Altafini, 2007b]), in that most of the critical points, for instance, are unstable but *not* repulsive.

For generic stationary target states, it can be shown explicitly that all of the unstable critical points except the global maximum in fact have attractive manifolds of positive dimension. For target states that are not stationary under the action of H_0 , there are additional complications in that the invariant set can be larger than the set of critical points of the Lyapunov function, although the set of target states $\rho_d(t)$ for which this happens in the ideal system case is of measure zero. Thus, while these issues complicate the problem for systems with strongly regular free (drift) Hamiltonian H_0 and fully connected control Hamiltonian H_1 , Lyapunov control is still generally effective in that for

most target states $\rho_d(t)$ the invariant set contains only the critical points of the Lyapunov function V , and the target state $\rho_d(t)$ is the only hyperbolic sink of the dynamical system.

The situation changes radically when either of the twin requirements of strong regularity of H_0 and full connectedness of H_1 are relaxed, even slightly. In this case the method becomes not only less effective, but the emergence of centre manifolds around the target state suggests that the method is likely to become ineffective in practice. Since the strict requirements above do not appear to be satisfied for most physical systems of interest, this suggests that the utility of this method for practical control field design is rather limited. It would be desirable to have stronger analytic results for non-stationary target states where well-established tools for stability and convergence analysis near fixed points are generally no longer applicable. Although some theoretical tools such as the Poincare map and Floquet's theorem [Perko, 2000] exist, to fully answer the question of stability and convergence of Lyapunov control for non-stationary (and in general non-periodic) target states, even for ideal bilinear Hamiltonian systems, would seem to require the development of new tools in dynamical systems theory.

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