OPTIMAL CONTROL OF MIXED-STATE QUANTUM SYSTEMS BASED ON LYAPUNOV METHOD

Shuang Cong, Yuanyuan Zhang
Department of Automation, University of Science and Technology of China, Hefei, Anhui, 230027, Republic of China

Kezhi Li
Dept.of Electrical and Electronic Engineering, Imperial College London, South Kensington Campus London SW7 2AZ, U.K.

Keywords: Optimal control, Lyapunov-based control, Quantum systems, Mixed state, Oscillator.

Abstract: An optimal control strategy of mixed state steering in finite-dimensional closed quantum systems is proposed in this paper. Two different situations are considered: one is the target state is in statistical incoherent mixtures of energy eigenstates in which the target states are diagonal. Another is not all of the off-diagonal elements in the target states are zeros. We change the trajectory tracking problem into the state steering one by introducing the unitary transformation with all energy eigenstates in the inner Hamiltonian of system controlled. Based on Lyapunov stability theorem the stable parameters of controller designed is selected and the optimality of the control law proposed is proven. Moreover, two numerical system control simulations are performed on the diatomic molecule described by the Morse oscillator model under the control law proposed. The system control simulation experimental results demonstrate that the control strategies proposed are efficient even when the controlled system is not completely controllable.

1 INTRODUCTION

As one of the greatest achievements in the 20th century, quantum mechanics has urged the human view of the matter to the microcosm. An enormous amount of revolution in theory and engineering science have been undertaken due to the development and applications of quantum physics, quantum chemistry, quantum computation and quantum information. (Nielsen and Chuang, 2000). In these new interdisciplinary fields, how to control the quantum systems has become a challenging subject. One part of the quantum control theories is about the applications of classical and modern control theory to quantum systems. (Wang and Schirmer, 2008) Now there have been various control schemes applied to the quantum systems, such as the Lyapunov-based method (Grivopoulos and Bamieh, 2003; Mirrahimi, Rouchon, and Turinici, 2005; Beauchard *et al.* 2007; Cong and Kuang, 2007; Kuang and Cong, 2008), optimal control method (Peirce, *et al.* 1988; D'Alessandro and Dahleh, 2001; Girardeau, *et al.* 1998; Schirmer, *et al.* 2000), learning control method (Judson and Rabitz, 1992; Phan and Rabitz, 1999), state estimation method (Doherty and Jacobs, 1999; Zhang, Li, and Guo, 2000), and stochastic control method (Belavkin, 1992; Bouten, *et al.* 2004), *etc.* Generally, the control aim of a quantum system is to search for a control field by means of minimizing an energy-type cost function of system that usually requires a maximal transition probability from an initial state to a particular target state. Among all of the quantum control strategies, optimal control methods are the most popular approaches that have been widely used specially in quantum chemistry fields. Since the mid 1980s, the quantum optimal control theory has attracted attentions from many researchers. However, many proposed optimal control methods are generally obtained by means of complex numeral iterative algorithms, which are off-line control methods and quite inconvenient to operate and realize. Thus, how to obtain an optimal method without iterative solutions is of great significance.

Zhang Y., Cong S. and Li K..
OPTIMAL CONTROL OF MIXED-STATE QUANTUM SYSTEMS BASED ON LYAPUNOV METHOD.
DOI: 10.5220/0003126000220030
In *Proceedings of the International Conference on Bio-inspired Systems and Signal Processing* (BIOSIGNALS-2011), pages 22-30
ISBN: 978-989-8425-35-5

There is another method called local optimal control which defines a general performance index $y(t)$ as a function of the expectation values of physical observables. Then it designs a control field that drives the quantum system satisfying the monotonous increasing condition of $y(t)$ (Ohtsuki, 1998; Sugawara, 2003). Local optimal law is explicitly derived without iteration and can satisfy the necessary condition for a solution to the optimal control problem. In the applications of optimal control theory, we can select different performance index and get different control laws. Here we will select the error between the states as a performance index of the control law. The difference between Ohtsuki (1998), Sugawara (2003) and this paper is that the derived control law in this paper satisfies the sufficient condition for optimal. We have applied this method to a pure state quantum system (Zhang and Cong, 2008). As a further step, in this paper we would like to consider the mixed-state control problem based on the formulas of statistical mechanics, and apply the idea in Ref. Zhang and Cong (2008) to the Liouville equation. In quantum system, two reasons lead to mixed-state: one is quantum dissipation due to quantum system entangle with environment. In such a situation, the system will be open. Quantum state will become a mixed-state even though it is a pure state at the beginning. Here, evolution of density matrix in this open system will not be unitary. Second, a mount of same particles in different pure states are incoherent mixed, which would be a quantum ensemble. Particles in different pure states are in this ensemble with some probability, viz. average statistically. In this paper, we only consider closed system without action with environment, so mixed-state here refers to mixed-state in ensemble.

The rest of the paper is organized as follows. In Sec. 2 we introduce the system models in Hilbert space and in Liouville space. Section 3 gives the control law theorem and its proof based on the Lyapunov stability theorem and principle of optimality under the condition that the target state is diagonal and non-diagonal, respectively. The numerical simulation on the diatomic molecule described by the Morse oscillator model is presented in Sec. 4. Finally, Sec. 5 concludes the study of the paper.

2 MODEL OF THE SYSTEM CONTROLLED

The state of a quantum mechanical system can be described in various ways. When a system is in a pure state un-entangled with its environment, the state of the system can be described by a wave function that evolves according to a control-dependent Schrödinger equation. One can also describe the state of the system by a density operator $\hat{\rho}(t)$, which can not only represent a pure state but also a mixed state. The density operator $\hat{\rho}(t)$ acting on the system's Hilbert space $\mathcal{H}$ evolves with time according to the quantum Liouville equation:

$$\begin{aligned} i\hbar\frac{\partial}{\partial t}\hat{\rho}(t) &= [\hat{H}(t), \hat{\rho}(t)], \\ \hat{H}(t) &= \hat{H}_0 + \sum_{m=1}^{M} f_m(t)\hat{H}_m \end{aligned} \tag{1}$$

where $\hat{H}_0$ is the system's internal (or free) Hamiltonian, and $\hat{H}_m$ is the interaction (or control) Hamiltonian, respectively, all of them will be assumed to be time-independent. $f_m(t)$ is the admissible real-valued external control field. We set the Planck constant $\hbar = 1$ for convenience.

Because $\hat{\rho}(t)$ is a $N \times N$ density matrix in Hilbert space, it's difficult to solve the differential Eq. (1). One may introduce the Liouville operator in the Liouville space according to the concept of Dirac operator to simplify this problem. There is a natural connection between the density matrix and Liouville space (Barnett and Dalton, 1987; Ohtsuki, *et al.* 1989). In the Liouville space, Eq. (1) can be represented in the same form as the Schrödinger equation

$$\begin{aligned} i\frac{\partial}{\partial t}\left|\rho(t)\right\rangle\!\right\rangle &= \mathcal{L}(t)\left|\rho(t)\right\rangle\!\right\rangle, \\ \mathcal{L}(t) &= \mathcal{L}_0 + \sum_{m=1}^{M} f_m(t)\mathcal{L}_m \end{aligned} \tag{2}$$

where $\left|\rho(t)\right\rangle\!\right\rangle$ is defined as a Liouville ket, and $\mathcal{L}$ is the Liouville operator defined by the dual correspondence

$$\mathcal{L}(t)\left|\rho(t)\right\rangle\!\right\rangle \leftrightarrow [\hat{H}, \hat{\rho}(t)] \tag{3}$$

The basis vectors respectively belonging to Liouville space and Hilbert space are defined by the

bijective correspondence $|mn\rangle\rangle \leftrightarrow |m\rangle\langle n|$ (Schirmer, 2000). Then one has

$$\begin{aligned}\mathcal{L}_{jk,mn} &= \langle\langle jk|\mathcal{L}|mn\rangle\rangle = \langle j|[\hat{H},|m\rangle\langle n|]|k\rangle \\ &= tr(|k\rangle\langle j|[\hat{H},|m\rangle\langle n|]) \\ &= \sum_i (\langle i||k\rangle\langle j|\hat{H}|m\rangle\langle n||i\rangle - \langle i||k\rangle\langle j||m\rangle\langle n|\hat{H}|i\rangle) \\ &= \langle j|\hat{H}|m\rangle\delta_{nk} - \langle n|\hat{H}|k\rangle\delta_{jm} = H_{jm}\delta_{nk} - H^*_{kn}\delta_{jm}\end{aligned} \quad (4)$$

For an $N \times N$ density matrix $\hat{\rho}(t)$ in Hilbert space, its replacement form $|\rho(t)\rangle\rangle$ is an N^2 column vector in Liouville space, and $\mathcal{L}$ is an $N^2 \times N^2$ matrix. In such a way it is much easier to solve Eq. (2) than Eq. (1) expressed in terms of some commutators. Hence, Eq. (2) will be adopted as the investigated model in following sections of the paper.

3 CONTROL LAW DESIGN

The quantum control problems can be formulated in state steering (or transfer) problem, that is to say steer the system from a given initial state to a desired target state. In this section we'll develop an optimal control method based on Lyapunov theorem for the Liouville equation.

First we will introduce principle of optimality and the sufficient condition for optimality. Suppose the controlled system is in the form of $\dot{\mathbf{x}}(t) = \mathbf{f}[\mathbf{x}(t), \mathbf{u}(t), t]$, where $t \in [T_1, T_2]$, $\mathbf{x}(t) \subset \mathbb{R}^n \times [T_1, T_2]$, $\mathbf{u}(t) \subset \mathbb{R}^m \times [T_1, T_2]$. Let X be a given region in $\mathbb{R}^n \times [T_1, T_2]$ and contain the target set S. For each $(\mathbf{x}_0, t_0)$ in X, one need determine the control $\mathbf{u}$ which transfers $(\mathbf{x}_0, t_0)$ to S and minimizes the performance index $J(\mathbf{x}, \mathbf{u}, t) = \int_{t_0}^{t_1} L[\mathbf{x}(t), \mathbf{u}(t), t]\mathrm{d}t$. Define $J^*(\mathbf{x}, t)$ is the minimum of $J(\mathbf{x}, \mathbf{u}, t)$. The Hamiltonian $H(\mathbf{x}, \mathbf{p}, \mathbf{u}, t)$ is given by

$$H(\mathbf{x}, \mathbf{p}, \mathbf{u}, t) = L(\mathbf{x}, \mathbf{u}, t) + \langle \mathbf{p}, \mathbf{f}(\mathbf{x}, \mathbf{u}, t) \rangle$$

Principle of Optimality: If $\mathbf{u}^*(t)$ is an optimal control and if $\mathbf{x}^*(t)$, for $t \in [t_0, t_1]$, is the optimal trajectory corresponding to $\mathbf{u}^*(t)$, then the restriction of $\mathbf{u}^*(t)$ to a subinterval $[t, t_1]$ of $[t_0, t_1]$ is an optimal control for the initial pair $(\mathbf{x}^*(t), t)$.

Sufficient Condition for Optimality (Athans and Falb, 1966)**:** Suppose that $X = \mathbb{R}^n \times (T_1, T_2)$, H is normal relative to $\mathbb{R}^n \times (T_1, T_2)$, and $\mathbf{u}(\mathbf{x}, \mathbf{p}, t)$ is the H-minimal control relative to $\mathbb{R}^n \times (T_1, T_2)$. Let $\mathbf{u}^*(t)$ be an admissible control such that:

a. $\mathbf{u}^*(t)$ transfers $(\mathbf{x}_0, t_0)$ to S.

b. There is a solution $J^*(\mathbf{x}, t)$ of the Hamilton-Jacobi equation

$$\frac{\partial J}{\partial t}(\mathbf{x}, t) + H[\mathbf{x}, \frac{\partial J}{\partial \mathbf{x}}(\mathbf{x}, t), \mathbf{u}(\mathbf{x}, \frac{\partial J}{\partial \mathbf{x}}(\mathbf{x}, t), t), t] = 0$$

satisfying the boundary condition $J(\mathbf{x}, t) = 0$ for $(\mathbf{x}, t) \in S$, such that

$$\mathbf{u}^*(t) = (\mathbf{x}^*(t), \frac{\partial J^*}{\partial \mathbf{x}}(\mathbf{x}^*(t), t), t)$$

for t in (t_0, t_1).

Then $\mathbf{u}^*(t)$ is an optimal control.

3.1 Stationary Target States

Assume the target state is the statistical incoherent mixtures of energy eigenstates: $\hat{\rho}_f = \sum_{n=1}^{N} w_n |n\rangle\langle n|$, $\hat{\rho}_f$ is a stationary target state, e.g. $\hat{\rho}_f = |0\rangle\frac{1}{4}\langle 0| + |1\rangle\frac{3}{4}\langle 1| = \frac{1}{4}\begin{pmatrix} 1 & 0 \\ 0 & 3 \end{pmatrix}$. In this case, all of the off-diagonal elements in the target state are zeros. If so, the optimal control law is given by the following theorem 1.

Theorem 1. For the system defined in the Liouville space by Eq. (2), given the performance index

$$J = \frac{1}{2}\int_0^\infty \{\sum_{m=1}^{M} \frac{1}{r_m}[\mathrm{Im}(\langle\langle \rho - \rho_f | P\mathcal{L}_m | \rho \rangle\rangle)]^2 + \mathbf{f}(t)^{\mathrm{T}} R \mathbf{f}(t)\}\mathrm{d}t \quad (5)$$

where $\mathbf{f}(t) = [f_1(t)\ f_2(t) \cdots f_M(t)]^T$, R is a diagonal matrix with positive elements, $r_m > 0$, $(m = 1, 2, \cdots, M)$, and P is a positive definite symmetric matrix that satisfies the equation

$$P\mathcal{L}_0 - \mathcal{L}_0^{\dagger} P = 0 \quad (6)$$

Then there exists an optimal control law

$$f_m^* = -\frac{1}{r_m}\mathrm{Im}(\langle\langle \rho - \rho_f | P\mathcal{L}_m | \rho \rangle\rangle), \quad (m = 1, 2, \cdots, M) \quad (7)$$

such that the system (2) is stable and the performance index (5) is minimum.

In fact, according to the Lyapunov indirect stability theorem, P is a positive definite symmetric matrix that should satisfy Lyapunov equation $P(i\mathcal{L}_0)+(i\mathcal{L}_0)^{\dagger}P=-Q$. Because $\mathcal{L}_0$ is a linear Hermitian operator, whose eigenvalues are real. So $i\mathcal{L}_0$ is a skew Hermitian operator, whose eigenvalues are pure imaginary. Accordingly, $Q=0$, which results in the condition (6).

Proof: (1) Proof of stability

Select the following Lyapunov function

$$V(|\rho\rangle\rangle)=\frac{1}{2}\langle\langle\rho-\rho_f|P|\rho-\rho_f\rangle\rangle \tag{8}$$

where P is a positive definite symmetric matrix satisfying Eq. (6). The first-order time derivative of $V(|\rho\rangle\rangle)$ is

$$\dot{V}(|\rho\rangle\rangle)=\mathrm{Re}(\langle\langle\rho-\rho_f|P|\dot{\rho}\rangle\rangle) \tag{9}$$

Substituting Eq. (2) into Eq. (9) yields

$$\begin{aligned}\dot{V}(|\rho\rangle\rangle)=&\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_0|\rho\rangle\rangle)+\\&\sum_{m=1}^{M}f_m(t)\,\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)\end{aligned} \tag{10}$$

Since $P\mathcal{L}_0-\mathcal{L}_0^{\dagger}P=0$ and $\mathcal{L}_0|\rho_f\rangle\rangle=0$, $\mathrm{Im}(\langle\langle\rho-\rho_f|PL_0|\rho\rangle\rangle)=0$ holds. Hence, Eq. (10) can be re-written as

$$\dot{V}(|\rho\rangle\rangle)=\sum_{m=1}^{M}f_m(t)\,\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle) \tag{11}$$

Substituting control law (7) into Eq. (11) yields

$$\dot{V}(|\rho\rangle\rangle)=-\sum_{m=1}^{M}\frac{1}{r_m}[\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)]^2\le 0 \tag{12}$$

Thus, the system (2) is stable under the control law (7). Next we will prove this control law is optimal.

(2) Proof of optimality

a) The Sufficient Condition for Optimality says that if a system can be transferred from some initial state to a target set by applying admissible control, then an optimal control exists and may be found by determining the admissible control f_m^* that causes the system to reach the target set S. A description of the target set S is assumed to be known. So for the system (2) it now only remains that one needs to construct a proper target set S. Here we use the similar way we have proven in reference 8 to construct the target set S. In fact, in the Lyapunov-based control design, the Lyapunov function V can be seen as a target set S. So one can define the target set S is the Lyapunov function V by constructing an appropriate matrix P.

P is selected a positive definite symmetric matrix that satisfies the Eq. (6). At the same time, the eigenvectors with the largest eigenvalue are the maxima of V, the eigenvectors with the smallest eigenvalue are the minima and all others are saddle points. Then select the smallest eigenvalue of P is P_f with the corresponding target state ρ_f. In such a way, a target set S with a monotonic function and the target state as the minima value are constructed, in which the initial state can be transferred to the target state by the control law f_m^*.

b) From Eq. (7) and Eq. (12), we can get $J^*(|\rho\rangle\rangle,t)$ as following

$$\begin{aligned}&J^*(|\rho\rangle\rangle,t)\\&=\frac{1}{2}\int_t^{\infty}\{\sum_{m=1}^{M}\frac{1}{r_m}[\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)]^2+\mathbf{f}^*(t)^{\mathrm{T}}R\mathbf{f}^*(t)\}\mathrm{d}t\\&=\int_t^{\infty}\{\sum_{m=1}^{M}\frac{1}{r_m}[\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)]^2\}\mathrm{d}t\\&=-\int_t^{\infty}\dot{V}(|\rho\rangle\rangle)dt=V(|\rho\rangle\rangle)\end{aligned} \tag{13}$$

Thus, the Hamiltonian function of the system can be

$$\begin{aligned}&H(|\rho\rangle\rangle,\mathbf{f})\\&=L(|\rho\rangle\rangle,\mathbf{f})+\mathrm{Im}[(\frac{\partial V(|\rho\rangle\rangle)}{\partial|\rho\rangle\rangle})^{\dagger}(\mathcal{L}_0+\sum_{m=1}^{M}f_m(t)\mathcal{L}_m)|\rho\rangle\rangle]\end{aligned} \tag{14}$$

where

$$L=\frac{1}{2}\sum_{m=1}^{M}\frac{1}{r_m}[\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)]^2+\mathbf{f}(t)^{\mathrm{T}}R\mathbf{f}(t)$$

Because $\frac{\partial J^*}{\partial t}(|\rho\rangle\rangle,t)=0$, a part of the sufficient condition for optimality is

$$\min_{\mathbf{f}\in R^M}[H(|\rho\rangle\rangle,\mathbf{f})]=0 \tag{15}$$

From Eq. (14), one can obtain

$$\begin{aligned}H(|\rho(t)\rangle\rangle,\mathbf{f})=&\frac{1}{2}\sum_{m=1}^{M}\frac{1}{r_m}[\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)]^2+\mathbf{f}(t)^{\mathrm{T}}R\mathbf{f}(t)+\\&\mathrm{Im}(\langle\langle\rho-\rho_f|P(\mathcal{L}_0+\sum_{m=1}^{M}f_m(t)\mathcal{L}_m)|\rho\rangle\rangle)\\=&\frac{1}{2}\sum_{m=1}^{M}\frac{1}{r_m}[\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)]^2+\\&\sum_{m=1}^{M}r_m f_m^2(t)+\sum_{m=1}^{M}f_m(t)\,\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)\\=&\frac{1}{2}\sum_{m=1}^{M}\frac{1}{r_m}[\mathrm{Im}(\langle\langle\rho-\rho_f|P\mathcal{L}_m|\rho\rangle\rangle)+r_m f_m(t)]^2\ge 0\end{aligned} \tag{16}$$

Substituting Eq. (7) into Eq. (16) yields

$$H(\left|\rho(t)\right\rangle\rangle, \mathbf{f}^*) = 0$$

Thus, the control law (7) is optimal and minimizes the performance index (5). The proof of theorem 1 is completed.

The design steps of the optimal control law proposed based on *Theorem* 1 are as follows:

(1) Select the weighting on the control vector

$$R = \text{diag}(r_i),\ r_i > 0,\ i = 1, 2, \cdots, m$$

(2) Solve Eq. (6) for obtaining the positive define matrix P.

(3) Calculate the optimal stabilizing control law from (7).

3.2 Non-stationary Target States

If not all of the off-diagonal elements in the target state are zeros, which is also a case of a mixed-state, e.g.

$$\hat{\rho}_f = |1\rangle\frac{1}{2}\langle 1| + (\frac{\sqrt{2}}{2}|0\rangle + \frac{\sqrt{2}}{2}|1\rangle)\frac{1}{2}(\frac{\sqrt{2}}{2}\langle 0| + \frac{\sqrt{2}}{2}\langle 1|) = \frac{1}{4}\begin{pmatrix} 1 & 1 \\ 1 & 3 \end{pmatrix}$$

In this case, the target state $\hat{\rho}_f(t)$ is in fact not stationary which evolves under $\hat{H}_0$ according to the Liouville-von Neumann equation

$$i\frac{\partial}{\partial t}\hat{\rho}_f(t) = [\hat{H}_0, \hat{\rho}_f(t)] \tag{17}$$

Now the target state is a time-dependent function, and the control problem becomes a trajectory tracking problem. From the system control point of view, a trajectory tracking problem can be easily solved by translating it into the state steering problem. To do so, we first carry out the following unitary transformations

$$\hat{\rho}(t) = U(t)\tilde{\rho}U^{\dagger}(t) \tag{18}$$

And

$$\hat{\rho}_f(t) = U(t)\tilde{\rho}_f U^{\dagger}(t) \tag{19}$$

in which $\tilde{\rho}_f$ is a stationary target state which equals $U(t) = diag(e^{-iE_1 t}, e^{-iE_2 t}, \cdots, e^{-iE_N t})$ and $E_i, i = 1, ..., N$ satisfy $\hat{H}_0 = diag(E_1, E_2, \cdots, E_N)$ in Eq. (1).

Substituting Eq. (18) into Eq. (1), one can obtain

$$i\frac{\partial}{\partial t}\tilde{\rho}(t) = [\sum_{m=1}^{M} f_m(t)\tilde{H}_m(t), \tilde{\rho}(t)] \tag{20}$$

where $\tilde{H}_m(t) = U^{+}(t)\hat{H}_m U(t)$.

Owing to the unitary transformation, $\hat{\rho}(t)$ and $\tilde{\rho}(t)$ have the same populations, which means that Eq. (1) and Eq. (20) describe the same physical system. In such a way, the problem of system (1) tracking a time-dependent target state $\hat{\rho}_f(t)$ in Eq. (17) is equivalent to a problem of steering the state in system (20) to the stationary target state $\tilde{\rho}_f$.

In the Liouville space, Eq. (20) can be represented as

$$i\frac{\partial}{\partial t}\left|\tilde{\rho}(t)\right\rangle\rangle = \sum_{m=1}^{M} f_m(t)\tilde{\mathcal{L}}_m(t)\left|\tilde{\rho}(t)\right\rangle\rangle \tag{21}$$

The optimal control law of Eq. (21) is given by the following theorem 2.

Theorem 2. For the system defined by Eq. (21), give the performance index

$$J = \frac{1}{2}\int_0^{\infty}\{\sum_{m=1}^{M}\frac{1}{r_m}[\text{Im}(\langle\langle\tilde{\rho} - \tilde{\rho}_f\left|P\tilde{\mathcal{L}}_m(t)\right|\tilde{\rho}\rangle\rangle)]^2 + \mathbf{f}(t)^{\mathrm{T}}R\mathbf{f}(t)\}\mathrm{d}t \tag{22}$$

where $\mathbf{f}(t) = [f_1(t)\ f_2(t)\cdots f_M(t)]^T$, R is a diagonal matrix with positive elements, $r_m > 0$, $(m = 1, 2, \cdots, M)$, and P is a positive definite symmetric matrix. Then there exists an optimal control law

$$f_m^* = -\frac{1}{r_m}\text{Im}(\langle\langle\tilde{\rho} - \tilde{\rho}_f\left|P\tilde{\mathcal{L}}_m(t)\right|\tilde{\rho}\rangle\rangle),\quad (m = 1, 2, \cdots, M) \tag{23}$$

such that the system (21) is stable and the performance index (22) is minimum.

The proof method of *theorem* 2 is the same as that of *theorem* 1, thus it will not be repeated here. In computer simulation, we need to choose an appropriate discrete propagation method to solve the differential equation (2) or (21). A simple approach would be adopting the first-order Euler method. But to obtain more efficient result, we employ four-order Runge-Kutta method, which has higher precision and faster convergence rate.

4 NUMERICAL SIMULATIONS AND RESULTS ANALYSIS

As an explicit example we consider a typical diatomic molecule model with N discrete

vibrational energy levels E_n corresponding to independent states $|n\rangle$ of the system. The internal Hamiltonian is given by

$$\hat{H}_0 = \sum_{n=1}^{N} E_n |n\rangle\langle n| \tag{24}$$

Assume that the diatomic molecular system is controlled by a single control field $f(t)$. Then the total Hamiltonian of the system can be represented as $\hat{H}(t) = \hat{H}_0 + f(t)\hat{H}_1$, and the corresponding Liouville operator is $\hat{\mathcal{L}}(t) = \hat{\mathcal{L}}_0 + f(t)\hat{\mathcal{L}}_1$. The interaction Hamiltonian can be chosen as the dipole form

$$\hat{H}_1 = \sum_{n=1}^{N-1} d_n (|n\rangle\langle n+1| + |n+1\rangle\langle n|) \tag{25}$$

Next we will separately study the diatomic molecules described by the Morse oscillator model and the Harmonic oscillator model.

4.1 Morse Oscillator Model

To simplify the calculation, we consider a hydrogen fluoride (HF) molecule described by a four-level Morse oscillator model. The vibrational energy levels are as follows (Schirmer, etc. 2001)

$$E_n = \hbar\omega_0 \left(n - \frac{1}{2}\right)\left[1 - \frac{1}{2}\left(n - \frac{1}{2}\right)B\right] \tag{26}$$

where $\omega_0 = 7.8\times10^{-14}\ \mathrm{s}^{-1}$ and $B = 0.0419$. Thus the corresponding energy levels are $E_1 = 0.4948$, $E_2 = 1.4529$, $E_3 = 2.3691$ and $E_4 = 3.2434$ in units of $\hbar\omega_0$. In the following calculations, all the parameters are expressed in atomic units (a.u.). Here the dipole moments in Eq. (25) are $d_n = \sqrt{n}$, $(n = 1,2,3)$. This system is completely controllable verified in Ref. Schirmer, *etc.* 2001.

Assume that the system is initially in the thermal equilibrium, i.e., $\hat{\rho}_0 = \sum_{n=1}^{4} w_n |n\rangle\langle n|$ with weights $w_n = C\exp[-E_n/(E_4 - E_1)]$. This is a Boltzmann distribution, and the normalization constant $C = (e^{-E_1/kT} + e^{-E_2/kT} + e^{-E_3/kT} + e^{-E_4/kT})^{-1}$ with $kT = E_4 - E_1$. Concretely, $w_1 = 0.3877$, $w_2 = 0.2736$, $w_3 = 0.1961$, and $w_4 = 0.1426$. The control task is to determine the control field $f(t)$ so as to steer the system from the initial state $\hat{\rho}_0$ to the target state $\hat{\rho}_f = \sum_{n=1}^{4} w_{5-n} |n\rangle\langle n|$. The state control problem and the observable control problem are inter-convertible. Thus the problem in this paper is equivalent to that in Refs. 13 and 14 with the goal to maximize the expectation value of the observable $\hat{A} = \hat{H}_0$.

According to *theorem* 1, the optimal control law can be obtained as

$$f(t) = -\frac{1}{r_1}\mathrm{Im}(\langle\langle \rho - \rho_f | P\mathcal{L}_1 | \rho \rangle\rangle) \tag{27}$$

The initial state of the system lies within the set of states resulting in $\mathrm{Im}(\langle\langle \rho_0 - \rho_f | P\mathcal{L}_1 | \rho_0 \rangle\rangle) = 0$, at the moment the control field $f_0 = 0$. This problem can be solved by applying an initial small magnitude disturbance to excite the system out of its initial equilibrium state (Beauchard, *et al.* 2007). In our numerical system simulations, the initial control field $f_0 = 0.05$ a.u., the target time $t_f = 200 a.u.$, and the sampling time $dt = 0.1$ a.u.. The suitable choice of the parameters r_1 and P is crucial to get good results. In order to obtain a higher probability of the target state, P can be chosen to make the Lyapunov function described by Eq. (8) larger at the initial time, and the diagonal elements of the initial state are ordered in a non-increasing sequence, the corresponding elements of P are also arrayed in non-increasing sequence (Kuang and Cong, 2008). After several times of tuning, we select $r_1 = 1$ and

$$P = diag(18,1,1,1,1,1.5,1,1,1,1,1,1,1,1,0.01)$$

The numerical simulation results are shown in Figures 1 to 4, in which Figure 1 shows the control field. The corresponding evolution populations of energy levels 1 through 4 are shown in Figure 2, from which one can see that the populations are inverted, i.e., the most energetic state $|4\rangle$ has the highest population, and the second one has the second highest population, etc. The final populations of energy levels are 0.1547, 0.1927, 0.2680, and 0.3845, respectively. Figure 3 shows the performance index, and Figure 4 shows the distance from the target state. At the target time, the distance is $\|\hat{\rho} - \hat{\rho}_f\|^2 = 0.0034$, so that the mixed-state control is completed. In Ref. Schirmer, *et al.* (2000)

$\hat{A} = \hat{H}_0$, and at the target time $t_f = 200$ a.u. the expectation value $\langle \hat{A}(t_f) \rangle$ is 99% of the theoretical maximum. While in this paper, this ratio is also 99% . Under the condition that the simulation result is the same, the design process of the control law in this paper is easier than that in Ref. Schirmer, *et al.* (2000) which needs iteration. Also, by comparing the results, we can find that the inverted rate of the levels is faster here, that is because the initial control value is larger. In the real applications, the control value can be tuned according to the requirement.

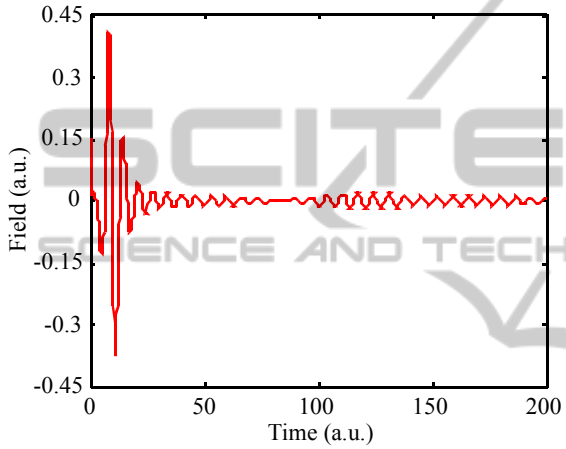

Figure 1: Optimal control field for a four-level Morse oscillator model.

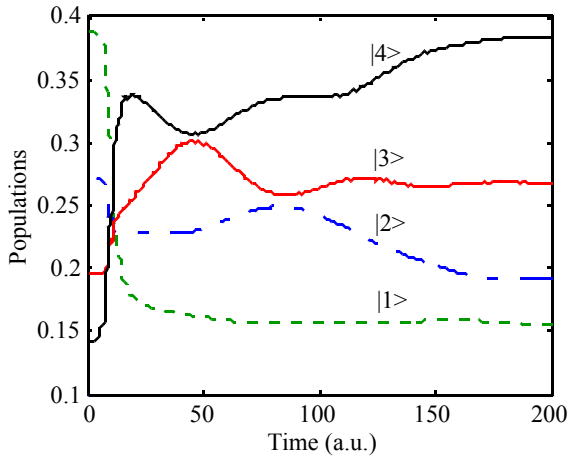

Figure 2: Evolution of populations for a four-level Morse oscillator model.

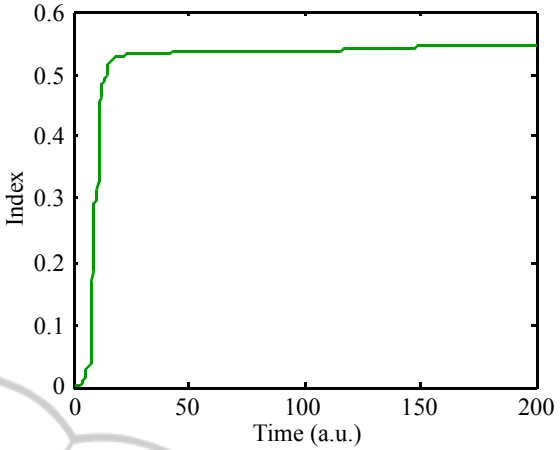

Figure 3: Performance index for a four-level Morse oscillator model.

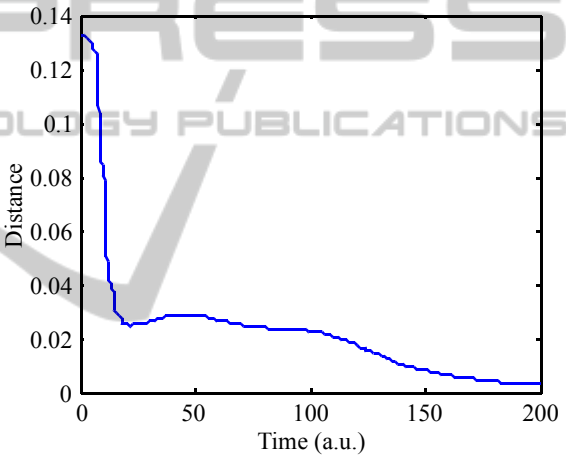

Figure 4: Distance from target state for a four-level Morse oscillator model.

4.2 Harmonic Oscillator Model

Comparing with the above mentioned completely controllable Morse oscillator model, here we consider the diatomic molecule described by a four-level Harmonic oscillator model. The vibrational energy levels are determined by

$$E_n = n - \frac{1}{2} \tag{28}$$

Thus the energy levels are $E_1 = 0.5$, $E_2 = 1.5$, $E_3 = 2.5$ and $E_4 = 3.5$. The dipole moments in this model are $d_n = 1$, $(n = 1,2,3)$. The system is not completely controllable because the dimension of the Lie algebra generated by $\hat{H}_0$ and $\hat{H}_1$ is less than 16 (Barnett and Dalton, 1987). We still suppose

that the initial density is $\hat{\rho}_0 = \sum_{n=1}^{4} w_n |n\rangle\langle n|$, in which $w_1 = 0.3850$, $w_2 = 0.2758$, $w_3 = 0.1976$ and $w_4 = 0.1416$. The target state and the control law are the same as that in the situation of the Morse oscillator model. Starting with $f_0 = 0.15$a.u., $dt = 0.1$a.u., $r_1 = 1$, and $P = diag(4,1,1,1,1,3,1,1,1,1,2,1,1,1,1,1)$, the simulation curves are shown in Figures 5-8. At the target time, the populations of energy levels are 0.1482, 0.2003, 0.2732, and 0.3783, respectively, and the distance from the target state is $\left\|\hat{\rho} - \hat{\rho}_f\right\|^2 = 0.0036$. Despite the system is not completely controllable, the method in this paper is still efficient.

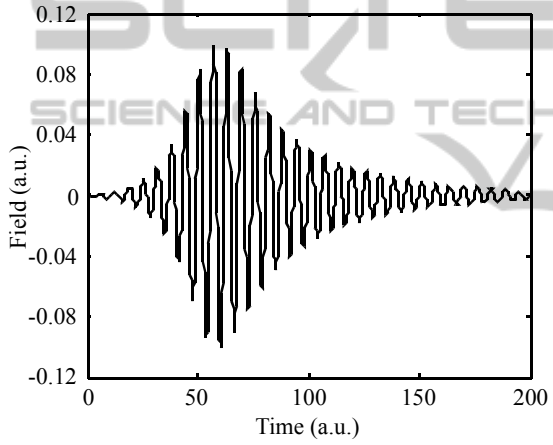

Figure 5: Optimal control field for a four-level Harmonic oscillator model.

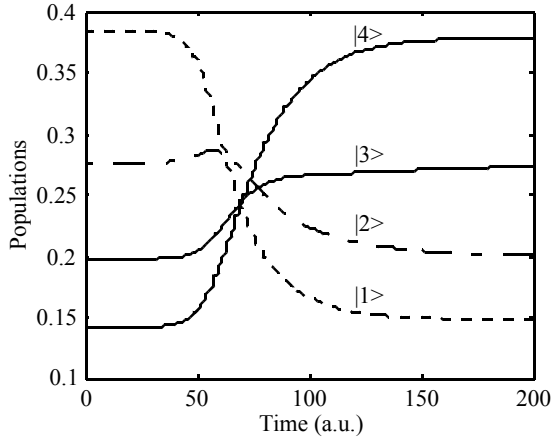

Figure 6: Evolution of populations for a four-level Harmonic oscillator model.

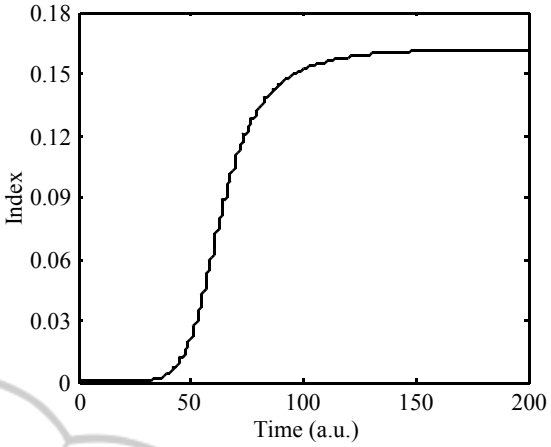

Figure 7: Distance from target state for a four-level Harmonic oscillator model.

5 CONCLUSIONS

In this paper we have developed an optimal control method based on Lyapunov theorem for the Liouville equation to realize the quantum control of the mixed states. The detailed design processes of the control laws have been given both in the cases of the target density operator of the system of interest being a diagonal form and a general one, respectively. Moreover, the numerical simulations were performed for the diatomic molecule described by the Morse oscillator model. The simulation results show that the method proposed is as efficient, even in the case that the system is not completely controllable.

ACKNOWLEDGEMENTS

This work was supported in part by the National Key Basic Research Program under Grants No. 2006CB922004 and No. 2009CB929601, the National Science Foundation of China under Grant No. 61074050.

REFERENCES

Athans, M. and Falb, P. L., 1966, Optimal Control *McGraw-Hill*, New York.

Barnett, S. M. and Dalton, B. J., 1987, Liouville space description of thermofields and their generalizations, *J. Phys.* A: Math. Gen. 20, 411-418.

Beauchard, K., Coron, J. M., Mirrahimi M. and Rouchon, P., 2007, Implicit Lyapunov control of finite dimensional Schrodinger equations, *Systems & Control Letters* 56, 388.

Belavkin, V. P., 1992, Quantum stochastic calculus and quantum nonlinear filtering, *Journal of Multivariate Analysis* 42, 171-201.

Bouten, L., Mâdâlin Guţâ, and Maassen, H., 2004, Stochastic Schrodinger equations, *J. Phys. A*, 37, 3189.

Cong, S. and Kuang, S., 2007, Quantum control strategy based on state distance, *Acta Automatica Sinica* 33, 28-31.

D'Alessandro, D. and Dahleh, M., 2001, Optimal control of two-level quantum systems, *IEEE Transactions on Automation Control*, 46, 866.

Doherty A. C.andJacobs, K., 1999, Feedback control of quantum systems using continuous state estimation, *Phys. Rev. A*, 60, 2700.

Girardeau, M. D. Schirmer, S. G. Leahy, J. V.and Koch, R. M., 1998, Kinematical bounds on optimization of observables for quantum systems, *Phys. Rev. A*, 58, 2684.

Grivopoulos, S. and Bamieh, B., 2003, Lyapunov-based control of quantum systems, in *Proceedings of the 42nd IEEE Conference on Decision and Control*, *Maui*, *Hawaii USA*.

Judson, R. S. and Rabitz, H., 1992, Teaching Lasers to Control Molecules, *Phys. Rev. Lett.* 68, 1500-1503.

Kuang, S. and Cong, S., 2008, Lyapunov control methods of closed quantum systems, *Automatica* 44, 98-108.

Mirrahimi, M. Rouchon, P. and Turinici, G., 2005, Lyapunov control of bilinear Schršdinger equations, *Automatica* 41: 1987.

Nielsen, M. A. and Chuang, I. L., 2000, *Quantum Computation and Quantum Information* (Cambridge University Press, England.

Ohtsuki, Y. and Fujimura, Y., 1989, *J. Chem. Phys.* 91, 3903.

Ohtsuki, Y. Kono, H. and Fujimura, Y., 1998, *J. Chem. Phys.*, 109, 9318.

Peirce, A. P. Dahleh, M. A. and Rabitz, H., 1988, Optimal control of quantum-mechanical systems: Existence, numerical approximation, and applications, *Phys. Rev. A,* 37, 4950 .

Phan, M. Q.and Rabitz, H., 1999, A self-guided algorithm for learning control of quantum-mechanical systems, *J. Chem. Phys.*, 110, 34 -41.

Schirmer, S. G., 2000, Ph.D. thesis, Oregon University.

Schirmer, S. G. Fu, H. and Solomon, A. I., 2001, Complete controllability of quantum systems, *Phys. Rev. A*, 63, 063410.

Schirmer, S. G., Girardeau, M. D. and Leahy, J. V., 2000, Efficient algorithm for optimal control of mixed-state quantum systems, *Phys. Rev. A*, 61, 012101.

Sugawara, M., 2003, General formulation of locally designed coherent control theory for quantum system J. Chem. Phys., 118, 6784-6800.

Wang, X. T. And Schirmer, S. G., 2008, Analysis of Lyapunov Method for Control of Quantum States, *quant-h/0801.0702.*

Zhang, C. W. Li, C. F. and Guo, G. C., 2000, Quantum clone and states estimation for n-state system, *Phys. Lett. A*, 271, 31-34.

Zhang, Y. Y. and Cong, S., 2008, Optimal quantum control based on the Lyapunov stability theorem, *Journal of University of Science and Technology of China* 38, 331-336.