Population Control of Equilibrium States of Quantum Systems via Lyapunov Method

KUANG Sen[1] CONG Shuang[1,2]

Abstract This paper studies the population control problem associated with the equilibrium states of mixed-state quantum systems by using a Lyapunov function with degrees of freedom. The control laws are designed by ensuring the monotonicity of the Lyapunov function; main results on the largest invariant set in the sense of LaSalle are given; and the strict expression of any state in the largest invariant set is normally deduced in the framework of Bloch vectors. By analyzing the obtained largest invariant set and the Lyapunov function itself, this paper also discusses the determination problem of the degrees of freedom. Numerical simulation experiments on a three-level system show the validity of research results.

Key words Quantum system, population control, Bloch vector, Lyapunov function, invariant set

DOI 10.3724/SP.J.1004.2010.01257

In the field of quantum systems, state control reduces to the so-called population control, when one neglects the phases between any eigenvectors that generate any given target state. In fact, population control can be regarded as a particular state control and is of very fundamental importance, e.g., in quantum chemistry. So far, for the population control of quantum states, several different strategies have emerged, e.g., optimal control[1−2], adiabatic control[3], factorization techniques of unitary group[4], Lyapunov method[5−14], methods based on adaptive tracking[15] and system decomposition[16], and so on.

Compared with other methods, Lyapunov method has some merits: its design procedure is simpler, its solving process is easier, and the physical meaning is more intuitive. However, the LaSalle invariance principle included in this method can only guarantee the convergence of closed-loop trajectories to some of its invariant set, and not to some prescribed state in the set. For this problem, [7] achieved an asymptotical approaching to an eigenstate by analyzing asymptotical tracking to any reference trajectory and using quantum adiabatic theorem. Reference [9] proposed an implicit Lyapunov method to achieve an asymptotical tracking to some eigenstate.

Reference [11] proposed a simple but tentative idea to achieve a satisfying transition to any eigenstate by constructing the degrees of freedom contained in a Lyapunov function. Theoretical analysis and simulation experiments show that the idea is very flexible and that the control effects are often excellent as long as the degrees of freedom are suitably constructed. Based on this, we further explore its application to mixed-state quantum systems. It is well known that equilibrium states are very important in system theory. For quantum systems described by pure states, equilibrium states in a strict sense do not exist since global phases evolve at each instant. However, equilibrium points in the sense of population still make sense. This implies that quantum systems described by Liouville equations contain true equilibrium states. This paper will focus on studying the population control of the equilibrium states associated with the Liouville equations.

Notice that the designed control laws in this paper contain the system states. To avoid notional confusion, it is necessary to clarify the difference of the terms "feedback control" and "closed-loop system" from the counterparts in the classical case. In fact, since the feedback states in the quantum field cannot usually be fully obtained by exact measurement, the designed control laws have the characteristics of open-loop implementation. This admits one to call such a control strategy "program control with state feedback"[11]. In terms of classical control theory, we can also call it "model-based feedback control", and the corresponding system "model-based closed-loop system". This is exactly the real meaning of the terms in this paper. Even so, the research in this paper still has theoretical and actual interests as, presently, open-loop quantum control remains important.

Manuscript received November 23, 2009; accepted March 17, 2010
Supported by National Key Basic Research Program of China (973 Program) (2006CB922004), National Natural Science Foundation of China (60904033, 60774098), the Chinese Postdoctoral Science Foundation (20100470848), and K. C. Wong Education Foundation, Hong Kong
1. Department of Automation, University of Science and Technology of China, Hefei 230027, P. R. China 2. Hefei National Laboratory for Physical Sciences at the Microscale, Hefei 230026, P. R. China

1 Preliminary notions and control design

1.1 Preliminary notions

This paper studies the following N-level quantum system and assumes that it is operator controllable:

$$\dot{\rho}(t) = -\mathrm{i}\left[H_0 + \sum_{k=1}^{m} H_k u_k(t),\ \rho(t)\right],\ \rho(0) = \rho_0 \qquad (1)$$

where $\rho(t)$ is the density operator describing the system dynamics; H_0, is the inner Hamiltonian of the system; $u_k(t)$ is an applied real-valued control field; H_k is the control Hamiltonian caused by the interaction between $u_k(t)$ and the system. Both H_0, and H_k are independent of time. We work in an orthonormal basis of energy eigenvectors. So, ρ, H_0, and H_k will take on the corresponding $N \times N$ matrix forms, and H_0 is diagonal.

The system described by (1) is closed. Its evolution is unitary. So, whatever value $u_k(t)$ takes, $\rho(t)$ and ρ_0 have the same spectrums. This is a necessary condition satisfied by any reachable state of ρ_0.

In physics, the eigenvalues λ_j $(j = 1, 2, \cdots, N)$ of $H_0 = \mathrm{diag}\{\lambda_1, \lambda_2, \cdots, \lambda_N\}$ represent all the possible energy values (energy levels) of the system, while $\omega_{jl} = \lambda_j - \lambda_l$ represents the Bohr frequency (transition frequency) between the energy levels λ_j and λ_l. Further, we give the following concepts. If all the energy levels of a quantum system are mutually different, then the system is called non-degenerate. If all the Bohr frequencies of a quantum system are mutually different, then the system is called transitionally non-degenerate. If there exists $k' \in \{1, 2, \cdots, m\}$ such that $(H_{k'})_{jl} \neq 0$ holds, then the energy-level pair (j, l) admits a direct transition and is called directly coupled. If for two arbitrary energy levels, there exists a path connecting them via a series of direct transitions, then the system is called connected. If two arbitrary energy levels admit a direct transition, then the system is called fully connected. Further, we call ρ satisfying $[H_0, \rho] = 0$ an equilibrium state of (1), denoted by ρ_e.

This paper assumes that the system under consideration is transitionally non-degenerate and connected.

1.2 Control design based on Lyapunov function

Our goal is to design the control laws $u_k(t)$ via the Lyapunov method such that the system (1) starts from an initial state ρ_0 and converges to (or transits with a high probability to) the population of some equilibrium state ρ_f isospectral to ρ_0.

Consider the Lyapunov function:

$$V(\rho) = \mathrm{tr}(P\rho) \tag{2}$$

where P is a positive definite Hermitian operator to be constructed and can be regarded as an imaginary mechanical quantity. Mathematically, $V(\rho)$ is a tracing operation, while physically, represents the average value of P. This Lyapunov function is a generalization of the counterpart in the pure state case in [11].

Now, design control laws by guaranteeing the monotonic decreasing of the Lyapunov function (2). Calculating its time derivative gives

$$\dot{V}(\rho) = -\mathrm{i}\,\mathrm{tr}([P, H_0]\rho) - \mathrm{i}\sum_{k=1}^{m} \mathrm{tr}([P, H_k]\rho)u_k \tag{3}$$

In view of the independence of the first item on the right-hand side of (3) from all the control components, let

$$[P, H_0] = 0 \tag{4}$$

To ensure $\dot{V}(\rho) \le 0$, one can design

$$u_k = \mathrm{i}\varepsilon_k \mathrm{tr}([P, H_k]\rho), \quad k = 1, \cdots, m \tag{5}$$

where $\varepsilon_k > 0$ and is used to adjust the amplitude of u_k.

Considering that the density operator ρ evolves in a unitary fashion, one can prove the following propositions about the Lyapunov function (2) and its extreme points via the tool of Lie algebra $su(N)$[17]:

Proposition 1. On the premise that ρ changes in a unitary fashion, if ρ is any extreme point of $V(\rho)$, then $[\rho, P] = 0$ holds; contrarily, if ρ satisfies $[\rho, P] = 0$, then ρ must be an extreme point of $V(\rho)$.

Further, one can also easily prove the following proposition about P:

Proposition 2. If H_0 is non-degenerate and satisfies $[H_0, P] = 0$, then P is diagonal.

Remark 1. The considered system in this paper is non-degenerate. So, by replacing P in Proposition 2 with ρ, it can be shown that its equilibrium states are also diagonal.

2 Main results on the largest invariant set

Since the system (1) with the control field (5) is autonomous, the LaSalle invariance principle can be used to analyze the convergence of the closed-loop system. This principle states that any trajectory of the closed-loop system must converge to the largest invariant set contained in the set $S = \{\rho : \dot{V}(\rho) = 0\}$.

Clearly, the set S is composed of all the extreme states of all the evolving processes (i.e., ρ such that $\dot{V}(\rho) = 0$). It is not hard to find from (3) $\sim$ (5) that S can be characterized by the following proposition.

Proposition 3. Assume that the closed-loop system (1) evolves to an extreme state $\rho(t_0)$ at time t_0. Then, the following three conditions are equivalent:

$$\dot{V}(\rho(t_0)) = 0 \tag{6}$$

$$\mathrm{tr}(\rho(t_0)[P, H_k]) = 0,\ k = 1, \cdots, m \tag{7}$$

$$u_k(t_0) = 0,\ k = 1, \cdots, m \tag{8}$$

Remark 2. Since (3) $\sim$ (5) do not involve concrete initial states, (6) $\sim$ (8) represent all the extreme states during the evolving processes starting from all the initial states. From $\mathrm{tr}(A[B, C]) = \mathrm{tr}(C[A, B])$, and Proposition 1 and (6), it is easily known that the extreme points of the Lyapunov function (2) with respect to ρ must be in S.

For the largest invariant set contained in S, we have the following theorem.

Theorem 1. Consider the closed-loop system (1) with the control field (5) and the following three conditions: 1) $[P, H_0] = 0$; 2) The system is non-degenerate, i.e., H_0 is a non-degenerate diagonal matrix; 3) The system is transitionally non-degenerate. Then, the following conclusions are true:

1) If Condition 1) holds, then the largest invariant set contained in S of the closed-loop system is $E = \{\rho(0) : \dot{V}(\rho(t)) = 0, t \in \mathbf{R}\}$, where $\rho(t)\,(t \in \mathbf{R})$ is the trajectory of the closed-loop system associated with the initial state $\rho(0)$.

2) If Conditions 1) and 2) simultaneously hold, then the largest invariant set in Conclusion 1) reduces to $E = \{\rho(0) : \mathrm{tr}(\mathrm{e}^{\mathrm{i}H_0 t} H_k \mathrm{e}^{-\mathrm{i}H_0 t}[\rho(0), P]) = 0, k = 1, \cdots, m; t \in \mathbf{R}\}$.

3) If Conditions 1) and 3) simultaneously hold, then the (l, j)-th element of the state $\rho(0)$ belonging to the largest invariant set in Conclusion 2) satisfies $(H_k)_{jl}(p_l - p_j)\rho_{lj}(0) = 0\,(j, l = 1, \cdots, N; k = 1, \cdots, m; j < l)$, where p_l and p_j are the l-th and j-th diagonal element of P, respectively.

Proof. See Appendix. □

Remark 3. The three conclusions in Theorem 1 cover three classes of cases about the system itself: Conclusion 1) imposes no restriction on the system; Conclusion 2) requires the system to be non-degenerate; while Conclusion 3) requires the system to be transitionally non-degenerate. It can be seen from the three conclusions that the expression of the states in the largest invariant set is more and more accessible to be analytically decided along with the gradual strengthening of the conditions on the system itself.

Remark 4. Actually, Conditions 2) and 3) are the conditions possessed by the system itself. However, the considered system in this paper satisfies Condition 3), and thereby Condition 2). That is, the largest invariant sets in the three conclusions of Theorem 1 are fully consistent for the system in this paper. Thus, from Proposition 1 and Conclusion 2) of Theorem 1, it follows that the extreme points of the Lyapunov function (2) with respect to ρ must also be in the largest invariant set E.

3 Bloch vector and convergent state set

Conclusion 3) of Theorem 1 shows the characteristics of each element of any state contained in the largest invariant set E. However, it is difficult to decide which states E is composed of on earth. In fact, given an initial state ρ_0, all the states which the closed-loop system may converge to are those contained in E and isospectral to ρ_0. We call the set of such states as the convergent state set of the closed-loop system, denoted by $E(\rho_0)$. To find $E(\rho_0)$ and achieve the convergence to the population of its some equilibrium state by constructing P, it is necessary to further search for the explicit expression of the states in E. To do this, the Bloch vector representation of density matrices will be introduced.

3.1 Bloch vector framework of density matrices

Assume that $\mathcal{H}_N$ is the Hilbert space associated with a N-level quantum system and isomorphic to $\mathbf{C}^N$. Then, the set of all the bounded linear operators that act on $\mathcal{H}_N$ and endowed with the following inner product forms a Hilbert space, called Liouville space, denoted by $\mathcal{L}(\mathcal{H}_N)$:

$$\langle\langle A|B\rangle\rangle = \mathrm{tr}(A^+B) \tag{9}$$

The inner product defined by (9) is also called the Hilbert-Schmidt inner product. With it, one can select the identity matrix I_N and the following generators of group $SU(N)$ as an orthonormal basis of $\mathcal{L}(\mathcal{H}_N)$[18−19]:

$$\sigma_{jl}^x = |j\rangle\langle l| + |l\rangle\langle j|,\ \ 1\le j<l\le N \tag{10}$$

$$\sigma_{jl}^y = -\mathrm{i}(|j\rangle\langle l| - |l\rangle\langle j|),\ \ 1\le j<l\le N \tag{11}$$

$$\sigma_j^z = \frac{\sqrt{2}}{\sqrt{j(j+1)}}\Big(\sum_{n=1}^{j}|n\rangle\langle n| - j|j+1\rangle\langle j+1|\Big),\quad 1\le j\le N-1 \tag{12}$$

where x, y, and z are used to distinguish different generators, and analogous to the Pauli matrices along the directions x, y, and z in the case of two-level systems. For convenience, this basis is written as

$$\{\sigma_s\}_{s=0}^{N^2-1} = \{I_N\}\cup\{\sigma_s\}_{s=1}^{N^2-1} = \{I_N\}\cup\{\sigma_{jl}^x,\sigma_{jl}^y,\sigma_j^z\} = \{I_N,\sigma_{jl}^x,\sigma_{jl}^y,\sigma_j^z\} \tag{13}$$

where $1\le j<l\le N$ and $1\le j\le N-1$.

The set of all the density matrices in $\mathcal{L}(\mathcal{H}_N)$ will form the so-called density matrix space, denoted by $\mathcal{L}_1(\mathcal{H}_N)$. Thus, any density matrix ρ in $\mathcal{L}_1(\mathcal{H}_N)$ can be expressed by the basis (13). In view of the intrinsic properties of density matrices, such an expression has a fixed form:

$$\rho = \frac{I_N}{N} + \frac{1}{2}\sum_{s=1}^{N^2-1}\gamma_s\sigma_s = \frac{I_N}{N} + \frac{1}{2}\sum_{j<l}\gamma_{jl}^x\sigma_{jl}^x + \frac{1}{2}\sum_{j<l}\gamma_{jl}^y\sigma_{jl}^y + \frac{1}{2}\sum_{j=1}^{N-1}\gamma_j^z\sigma_j^z \tag{14}$$

where $\gamma_s = \mathrm{tr}(\rho\sigma_s)\,(1\le s\le N^2-1)$, which can be verified by using (9) and the orthogonality of the basis (13).

The vector $\boldsymbol{\gamma} = [\gamma_1,\gamma_2,\cdots,\gamma_{N^2-1}]^{\mathrm{T}}$ is the Bloch vector of ρ. Clearly, it is a real-valued vector in $\mathbf{R}^{N^2-1}$. The set of all the Bloch vectors forms the Bloch space of the system in $\mathbf{R}^{N^2-1}$, denoted by $\mathcal{B}(\mathbf{R}^{N^2-1})$. In general, given some members of the basis (13), one cannot generate ρ by taking arbitrary $\boldsymbol{\gamma}$ in (14). Alternatively, it should be obtained by taking $\boldsymbol{\gamma}$ in the Bloch space $\mathcal{B}(\mathbf{R}^{N^2-1})$. For any N-level quantum system, [19] proved the following proposition on its Bloch space:

Proposition 4. Let $a_\nu(\boldsymbol{\gamma})$ be the coefficients of the characteristic polynomial $\det(\eta I_N - \rho)$ with respect to ρ in (14), and define $\mathcal{B}(\mathbf{R}^{N^2-1}) = \{\boldsymbol{\gamma}\in\mathbf{R}^{N^2-1} : a_\nu(\boldsymbol{\gamma})\ge 0\,(\nu=1,\cdots,N)\}$. Then, the map: $\boldsymbol{\gamma}\in\mathcal{B}(\mathbf{R}^{N^2-1}) \longrightarrow \rho = \frac{1}{N}I_N + \frac{1}{2}\sum_{s=1}^{N^2-1}\gamma_s\sigma_s \in \mathcal{L}_1(\mathcal{H}_N)$ is a bijection from the Bloch space $\mathcal{B}(\mathbf{R}^{N^2-1})$ to the density matrix space $\mathcal{L}_1(\mathcal{H}_N)$.

Proposition 4 is important in theory, as it directly indicates a one-to-one correspondence between the density matrix space and the Bloch space of any N-level system. Particularly, it can be calculated from Proposition 4 that the Bloch space of any two-level system is a well-known unit ball $\mathcal{B}(\mathbf{R}^3) = \{\boldsymbol{\gamma}\in\mathbf{R}^3 : |\boldsymbol{\gamma}|\le 1\}$ in $\mathbf{R}^3$.

3.2 Convergent state set of the closed-loop system

Conclusion 3) of Theorem 1 does not cover the case "$j=l$". In fact, for $j=l$, the expression in Conclusion 3) naturally holds. So, all the basis members of diagonal type, $\sigma_j^z\,(j=1,\cdots,N-1)$, will become the generators of the largest invariant set E. For $j\ne l$, it can be known from the expression in Conclusion 3) that, if at least one of the following two conditions is satisfied:

$$\exists\, j',l'\in\{1,\cdots,N\},\ \text{s.t. } p_{l'} = p_{j'},\ j'<l' \tag{15}$$

and

$$\exists\, j',l'\in\{1,\cdots,N\},\ \text{s.t. } (H_k)_{j'l'} = 0,\quad k=1,\cdots,m,\ j'<l' \tag{16}$$

then

$$\rho_{l'j'}(0)\in\mathbf{C},\ j'<l' \tag{17}$$

Equation (17) means that the basis members $\sigma_{j'l'}^x$ and $\sigma_{j'l'}^y$ are also the generators of E. Generally, when the control Hamiltonians $H_k\,(k=1,\cdots,m)$ are given and P is determined beforehand, it is not difficult to find all the pairs of the basis members acting as the generators of E. Thus, E can be ultimately calculated via (14). Accordingly, the following theorem is obtained.

Theorem 2. Consider the transitionally non-degenerate system (1) with the control field (5). If $[P,H_0]=0$, then any trajectory of the closed-loop system converges to the largest invariant set contained in S, $E = \{\rho : \rho = \frac{1}{N}I_N + \frac{1}{2}\sum_{j=1}^{N-1}\gamma_j^z\sigma_j^z + \frac{1}{2}\sum_{j<l}\gamma_{jl}^x\sigma_{jl}^x + \frac{1}{2}\sum_{j<l}\gamma_{jl}^y\sigma_{jl}^y,\ ((H_k)_{jl} = 0, k=1,\cdots,m, \text{or } p_j = p_l; \boldsymbol{\gamma}\in\mathcal{B}(\mathbf{R}^{N^2-1}))\}$, where $\boldsymbol{\gamma}$ is the Bloch vector of dimension N^2-1 with γ_{jl}^x, γ_{jl}^y and $\gamma_j^z\,(j=1,\cdots,N; j<l)$ as components and satisfies that γ_{jl}^x and $\gamma_{jl}^y\,(j=1,\cdots,N; j<l)$ associated with $(H_k)_{jl}\ne 0\,(k\in 1,\cdots,m)$ are equal to 0.

Since the equilibrium states of the considered systems in this paper are diagonal matrices, the set of all the equilibrium states which the closed-loop system may converge to is the set of all the diagonal matrices in the largest invariant set E, denoted by E_e. Further, given an initial state ρ_0, the set of all the equilibrium states which the closed-loop system may converge to is the set of all the diagonal matrices in the convergent state set $E(\rho_0)$, denoted by $E_e(\rho_0)$. Evidently, the number of the elements in $E_e(\rho_0)$ is finite, denoted by ρ_{e1}, ρ_{e2}, $\cdots$, ρ_{en} $(n\le N!)$.

Note that when P is non-degenerate and the controlled system is fully connected, the following corollary is easily obtained via Theorem 2, Propositions 1 and 2 (see also [17]).

Corollary 1. For the transitionally non-degenerate system (1) with the control field (5), if P is non-degenerate and the system (1) is fully connected, then the largest invariant set in Theorem 2 reduces to the equilibrium state set E_e, that is, any trajectory of the closed-loop system converges to one of the equilibrium states of the system (1).

4 Determination of degrees of freedom

4.1 Considerations on the determination of *P*

In this section, the determination problem of P will be studied so that the system (1) can converge to (or transit with a high probability to) the population of its some equilibrium state. This task is not so formidable because the system only needs to be steered to a state that has the same population as the target equilibrium state.

According to Theorem 2, the largest invariant set E depends not only on the pairs of the energy levels that are not directly coupled in the control Hamiltonians but also on the diagonal elements of P. For the considered system in this paper, Theorem 2 and the diagonal type of the equilibrium states ensure that all the equilibrium states are in the largest invariant set. However, this does not mean that any closed-loop trajectory can furthest approach the population of the target equilibrium state. This paper solves this problem by constructing P.

In view of the diagonal type of P, (2) can be written as

$$V(\rho)=\sum_{k=1}^{N}p_k\rho_{kk} \tag{18}$$

where ρ_{kk} is the (k,k)-th element of ρ, and represents the population component on the k-th eigenstate. Equation (18) and the population conservation during the evolving process ensure that the population components on the eigenstates associated with the maximal and minimal diagonal elements of P changes quickly with the decreasing of V. Based on this, the diagonal element of P associated with the maximal diagonal element of the target equilibrium state should be kept minimal; the diagonal element of P associated with the minimal diagonal element of the target equilibrium state should be kept maximal; while other diagonal elements should suitably take values.

Such an approach of roughly determining diagonal elements cannot guarantee the utmost approach to the population of the target equilibrium state. So, it is necessary to further adjust those diagonal elements. An idea is to observe and alter the diagonal values of P via simulation experiments to resultantly alter the changing rate of the Lyapunov function (2) with respect to the population components on the eigenstates. Furthermore, altering the changing rate of the Lyapunov function (2) with respect to time can also alter the closed-loop trajectory. Considering (3) $\sim$ (5), one can obtain

$$\dot V(\rho)=-4\sum_{k=1}^{m}\varepsilon_k\Big(\sum_{j<l}(p_j-p_l)\Im(\rho_{jl}(H_k)_{lj})\Big)^2 \tag{19}$$

It can be seen from (19) that the pairs of the energy levels that are directly coupled in the control Hamiltonians decide the diagonal elements of P that impact on the changing rate of the Lyapunov function (2). So, adjusting the differences between the diagonal elements of P associated with the energy levels that are directly coupled can alter the decreasing rate of the Lyapunov function (2).

4.2 Illustrative example

This subsection will expatiate on the concrete determination method of P via a numerical example. Consider a three-level system influenced by only one control field. In the orthonormal basis $\{|0\rangle=[1,0,0]^{\rm T},|1\rangle=[0,1,0]^{\rm T},|2\rangle=[0,0,1]^{\rm T}\}$, the inner and control Hamiltonians are given as $H_0=\begin{bmatrix}0.3&0&0\\0&0.5&0\\0&0&0.9\end{bmatrix}$ and $H_1=\begin{bmatrix}0&1&0\\1&0&1\\0&1&0\end{bmatrix}$, respectively. Assume that this system is initially in the state $|\psi_1(0)\rangle=|0\rangle$ with probability 0.9 and the state $|\psi_2(0)\rangle=\frac{1}{\sqrt2}|0\rangle+\frac{\rm i}{2}|1\rangle+\frac{\rm i}{2}|2\rangle$ with probability 0.1, that is, the initial density operator is $\rho(0)=\begin{bmatrix}0.95&\frac{(-{\rm i}\sqrt2)}{40}&\frac{(-{\rm i}\sqrt2)}{40}\\\frac{({\rm i}\sqrt2)}{40}&0.025&0.025\\\frac{({\rm i}\sqrt2)}{40}&0.025&0.025\end{bmatrix}$.

According to Theorem 2, it is easy to find the largest invariant set E and the convergent state set $E(\rho_0)$, where the Bloch space $\mathcal{B}(\mathbf{R}^8)$ can be calculated by using Proposition 4[19]. Also, numerical computation shows that the three eigenvalues of $\rho(0)$ are almost equal to 0, 0.0472, and 0.9528. That is to say, the set $E_e(\rho(0))$ of the equilibrium states to which the closed-loop system may converge contains 3! elements: $\rho_{e1}={\rm diag}\{0,0.0472,0.9528\}$, $\rho_{e2}={\rm diag}\{0,0.9528,0.0472\}$, $\rho_{e3}={\rm diag}\{0.0472,0,0.9528\}$, $\rho_{e4}={\rm diag}\{0.0472,0.9528,0\}$, $\rho_{e5}={\rm diag}\{0.9528,0,0.0472\}$, and $\rho_{e6}={\rm diag}\{0.9528,0.0472,0\}$.

The control goal is to drive this system to the population of ρ_{e1} (with a high probability), i.e., to let this system utmostly approach the quantum state with the population 0, 0.0472, and 0.9528 on the three eigenstates. Based on Subsection 4.1, the following parameters are selected in the simulation experiments: $p_1=1$, $p_2=0.7$, $p_3=0.5$, and $\varepsilon_1=0.05$ in the control field (5). Simulation results show that when the control period t_f is large enough (e.g., $t_f>10\,000$a.u.), the population on the three eigenstates finally reaches up to about 0.0003, 0.0472, and 0.9525, which is very close to the population on the three eigenstates of ρ_{e1}. To clearly see the key evolving process of the closed-loop system, the corresponding simulation curves in the time interval $[0,1\,000]$ are plotted in Figs. 1 and 2.

It can be seen from Figs. 1 and 2 that the population evolution of the system goes through the path $|1\rangle\rightarrow|2\rangle\rightarrow|3\rangle$. Evidently, the population exchange between the eigenstates $|1\rangle$ and $|2\rangle$ mainly happens in the time interval $[0,380]$, while the population exchange between $|2\rangle$ and $|3\rangle$ mainly happens in $[380,880]$. Numerical calculation shows that the oscillatory frequencies of the control field in $[0,380]$ and $[380,880]$ are about 0.1984 and 0.4021, which are very close to the transition frequencies 0.2 (between $|1\rangle$ and $|2\rangle$) and 0.4 (between $|2\rangle$ and $|3\rangle$), respectively.

In fact, the better control results can be obtained by suitably adjusting the diagonal elements of P. For instance, for $p_1=1.2$, $p_2=0.7$, and $p_3=0.3$, the population on the three eigenstates finally reaches up to about 0, 0.0472, and 0.9528. Further, it follows from (19) that the period which the system arrives at the steady population decreases as the decreasing rate of the Lyapunov function (2) increases.

5 Conclusion

For transitionally non-degenerate quantum systems, this paper has studied the convergence or high-probability transition to the population of some equilibrium state by using a Lyapunov function with degrees of freedom. Based on the LaSalle principle, the largest invariant set of the closed-loop systems has been analyzed. Particularly, the explicit expression of the states in the largest invariant set has been given in the framework of the Bloch vectors of density matrices. Also, we have discussed the determination problem of P so that the controlled system can utmostly approach the population of any target equilibrium state.

The simulation experiments on a three-level system have verified the effectiveness of the research results. However, when the number of the energy levels of the system is relatively large or the structure of the largest invariant set is very complex, the trial determination method in this paper appears weak. In this case, it is necessary to develop some new theoretical tools and further search for some more rigorous determination principles.

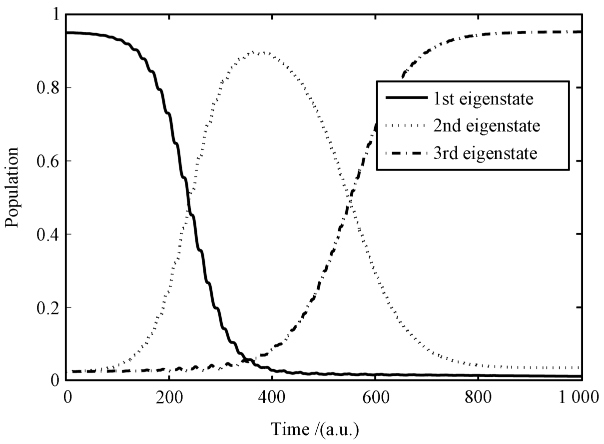

Fig. 1 The population evolving curves in the interval $[0, 1\,000]$

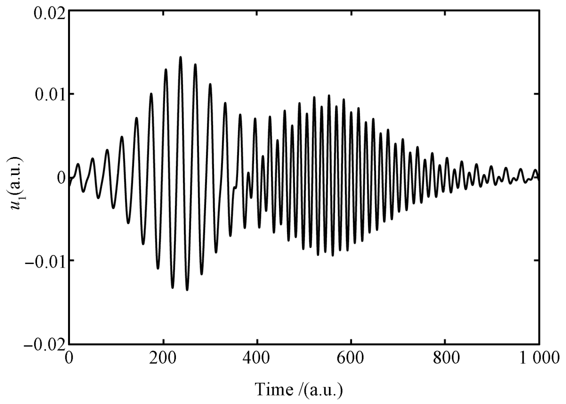

Fig. 2 The changing curve of the control field in $[0, 1\,000]$

Appendix The proof of Theorem 1

A.1 Conclusion 1)

It can be known from Proposition 3 that the states satisfying $\dot{V}(\rho(t)) = 0\,(t \in \mathbf{R})$ are the ones satisfying

$$u_k(t) = \mathrm{i}\varepsilon_k \mathrm{tr}([P, H_k]\rho(t)) = 0,\ k = 1, \cdots, m,\ t \in \mathbf{R} \tag{A1}$$

This implies that $\rho(t)$ contained in the control field is the free evolving state of the system. Substituting the solution of $\dot{\rho}(t) = -\mathrm{i}[H_0, \rho(t)]$ into (A1) equivalently gives

$$\mathrm{tr}(\mathrm{e}^{-\mathrm{i}H_0 t}\rho(0)\mathrm{e}^{\mathrm{i}H_0 t}[P, H_k]) = 0,\ k = 1, \cdots, m,\ t \in \mathbf{R} \tag{A2}$$

That is to say, the largest invariant set contained in S is

$$E = \{\rho(0) : \mathrm{tr}(\mathrm{e}^{-\mathrm{i}H_0 t}\rho(0)\mathrm{e}^{\mathrm{i}H_0 t}[P, H_k]) = 0,\ k = 1, \cdots, m,\ t \in \mathbf{R}\} \tag{A3}$$

Now, we use (A2) to prove the invariance and the largest property of E. Firstly, for showing the invariance, we suppose $\rho_1(0) \in E$, i.e.,

$$\mathrm{tr}(\mathrm{e}^{-\mathrm{i}H_0 t}\rho_1(0)\mathrm{e}^{\mathrm{i}H_0 t}[P, H_k]) = 0,\ k = 1, \cdots, m,\ t \in \mathbf{R} \tag{A4}$$

Then, the system state at any time t_0 is equal to $\rho_1(t_0) = \mathrm{e}^{-\mathrm{i}H_0 t_0}\rho_1(0)\mathrm{e}^{\mathrm{i}H_0 t_0}$. Let $\rho_1(t_0)$ be a new initial state, denoted by $\rho_1(t_0)(0)$. Calculating the left-hand side of (A2) gives

$$\mathrm{tr}(\mathrm{e}^{-\mathrm{i}H_0 t}\rho_1(t_0)(0)\mathrm{e}^{\mathrm{i}H_0 t}[P, H_k]) = \mathrm{tr}(\mathrm{e}^{-\mathrm{i}H_0 (t+t_0)}\rho_1(0)\mathrm{e}^{\mathrm{i}H_0 (t+t_0)}[P, H_k]) \tag{A5}$$

Clearly, for $t \in \mathbf{R}$, (A5) is equivalent to the left-hand side of (A4), i.e., $\rho_1(t_0) \in E$. This ends the proof of the invariance.

Next, for proving the largest invariant property of E, suppose E' is any invariant set contained in S and $\rho'(0)$ is any point in E'. The invariance of E' ensures that the trajectory starting from $\rho'(0)$ still belongs to E' and can be written as $\rho'(t) = \mathrm{e}^{-\mathrm{i}H_0 t}\rho'(0)\mathrm{e}^{\mathrm{i}H_0 t}\ (t \in \mathbf{R})$, i.e., $\rho'(0) = \mathrm{e}^{\mathrm{i}H_0 t}\rho'(t)\mathrm{e}^{-\mathrm{i}H_0 t}$. Replacing $\rho(0)$ in (A2) with $\rho'(0)$ gives $\mathrm{tr}(\rho'(t)[P, H_k]) = 0\,(k = 1, \cdots, m; t \in \mathbf{R})$. This is exactly the characteristic of E (see (A1)). The arbitrariness of $\rho'(0)$ in E' ensures that $E' \subset E$. Further, the arbitrariness of E' in S ensures that E is the largest invariant set contained in S.

A.2 Conclusion 2)

Since H_0 is non-degenerate and diagonal, it can be known from Proposition 2 that P is also diagonal. So, one has $P = \mathrm{e}^{-\mathrm{i}H_0 t}P\mathrm{e}^{\mathrm{i}H_0 t}$. Combined with $\mathrm{tr}(A[B, C]) = \mathrm{tr}(C[A, B])$, the left-hand side of (A2) can be reduced to

$$\begin{aligned}
&\mathrm{tr}(\mathrm{e}^{-\mathrm{i}H_0 t}\rho(0)\mathrm{e}^{\mathrm{i}H_0 t}[P, H_k]) = \\
&\quad \mathrm{tr}(H_k[\mathrm{e}^{-\mathrm{i}H_0 t}\rho(0)\mathrm{e}^{\mathrm{i}H_0 t}, P]) = \\
&\quad \mathrm{tr}(H_k[\mathrm{e}^{-\mathrm{i}H_0 t}\rho(0)\mathrm{e}^{\mathrm{i}H_0 t}, \mathrm{e}^{-\mathrm{i}H_0 t}P\mathrm{e}^{\mathrm{i}H_0 t}]) = \\
&\quad \mathrm{tr}(H_k \mathrm{e}^{-\mathrm{i}H_0 t}[\rho(0), P]\mathrm{e}^{\mathrm{i}H_0 t}) = \\
&\quad \mathrm{tr}(\mathrm{e}^{\mathrm{i}H_0 t}H_k \mathrm{e}^{-\mathrm{i}H_0 t}[\rho(0), P])
\end{aligned}$$

Thus, (A2) is equivalent to

$$\mathrm{tr}(\mathrm{e}^{\mathrm{i}H_0 t}H_k \mathrm{e}^{-\mathrm{i}H_0 t}[\rho(0), P]) = 0,\ k = 1, \cdots, m,\ t \in \mathbf{R} \tag{A6}$$

and accordingly, (A3) can be equivalently written as

$$E = \{\rho(0) : \mathrm{tr}(\mathrm{e}^{\mathrm{i}H_0 t}H_k \mathrm{e}^{-\mathrm{i}H_0 t}[\rho(0), P]) = 0,\ k = 1, \cdots, m,\ t \in \mathbf{R}\} \tag{A7}$$

A.3 Conclusion 3)

Applying $\mathrm{e}^{A}B\mathrm{e}^{-A} = \sum_{n=0}^{\infty} \frac{[A^{(n)}, B]}{n!}$ to (A6) gives

$$\begin{aligned}
&\mathrm{tr}\left(\mathrm{e}^{\mathrm{i}H_0 t}H_k \mathrm{e}^{-\mathrm{i}H_0 t}[\rho(0), P]\right) = 0 \Longleftrightarrow \\
&\quad \mathrm{tr}\left(\sum_{n=0}^{\infty}\frac{1}{n!}\left[(\mathrm{i}H_0 t)^{(n)}, H_k\right][\rho(0), P]\right) = 0 \Longleftrightarrow \\
&\quad \sum_{n=0}^{\infty}\frac{(\mathrm{i}^n t^n)}{n!}\mathrm{tr}\left(\left[H_0^{(n)}, H_k\right][\rho(0), P]\right) = 0
\end{aligned} \tag{A8}$$

$$M = \begin{bmatrix} 1 & \cdots & 1 & 1 & \cdots & 1 & \cdots & 1 \\ \omega_{12}^2 & \cdots & \omega_{1N}^2 & \omega_{23}^2 & \cdots & \omega_{2N}^2 & \cdots & \omega_{N-1,N}^2 \\ \omega_{12}^4 & \cdots & \omega_{1N}^4 & \omega_{23}^4 & \cdots & \omega_{2N}^4 & \cdots & \omega_{N-1,N}^4 \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \ddots & \vdots \\ \omega_{12}^{N(N-1)-2} & \cdots & \omega_{1N}^{N(N-1)-2} & \omega_{23}^{N(N-1)-2} & \cdots & \omega_{2N}^{N(N-1)-2} & \cdots & \omega_{N-1,N}^{N(N-1)-2} \end{bmatrix}$$

where $[H_0^{(n)}, H_k] = \underbrace{[H_0, [H_0, \cdots, [H_0}_{n \text{ times}}, H_k]]]$. Particularly, $[H_0^{(0)}, H_k] = H_k$.

By considering the linear independence of time sequence 1, t, t^2, $\cdots$, (A8) can be written as

$$\text{tr}\left([H_0^{(n)}, H_k][P, \rho(0)]\right) = 0, \quad n = 0, 1, 2, \cdots, \ k = 1, \cdots, m \tag{A9}$$

Denoting P as $P = \text{diag}\{p_1, p_2, \cdots, p_N\}$ and considering the diagonal type of H_0, one can calculate $[H_0^{(n)}, H_k]$ and $[P, \rho(0)]$ as

$$[H_0^{(n)}, H_k] = ((\lambda_j - \lambda_l)^n (H_k)_{jl}) = (\omega_{jl}^n (H_k)_{jl}), \quad j, l = 1, \cdots, N \tag{A10}$$

and

$$[P, \rho(0)] = ((p_j - p_l)\rho_{jl}(0)), \ j, l = 1, \cdots, N \tag{A11}$$

respectively.

Substituting (A10) and (A11) into (A9) gives

$$\sum_{j,l=1}^{N} \omega_{jl}^n (H_k)_{jl} (p_l - p_j) \rho_{lj}(0) = 0, \quad n = 0, 1, 2, \cdots, \ k = 1, \cdots, m \tag{A12}$$

By using the Hermitian property of H_k and $\rho(0)$, (A12) can be further written as

$$\sum_{j<l} \left(\omega_{jl}^n (H_k)_{jl} (p_l - p_j) \rho_{lj}(0) + \omega_{lj}^n (H_k)_{jl}^* (p_j - p_l) \rho_{lj}^*(0)\right) = 0 \tag{A13}$$

When n is even, (A13) can be reduced to

$$\sum_{j<l} \omega_{jl}^n \Im((H_k)_{jl} (p_l - p_j) \rho_{lj}(0)) = 0, \quad n = 0, 2, \cdots, \ k = 1, \cdots, m \tag{A14}$$

When n is odd, (A13) can be reduced to

$$\sum_{j<l} \omega_{jl}^n \Re((H_k)_{jl} (p_l - p_j) \rho_{lj}(0)) = 0, \quad n = 1, 3, \cdots, \ k = 1, \cdots, m \tag{A15}$$

Denote

$$\boldsymbol{\xi}_k = \begin{bmatrix} (H_k)_{12}(p_2 - p_1)\rho_{21}(0) \\ \vdots \\ (H_k)_{1N}(p_N - p_1)\rho_{N1}(0) \\ (H_k)_{23}(p_3 - p_2)\rho_{32}(0) \\ \vdots \\ (H_k)_{2N}(p_N - p_2)\rho_{N2}(0) \\ \vdots \\ (H_k)_{N-1,N}(p_N - p_{N-1})\rho_{N,N-1}(0) \end{bmatrix}$$

$$\Lambda = \text{diag}\{\omega_{12}, \cdots, \omega_{1N}, \omega_{23}, \cdots, \omega_{2N}, \cdots, \omega_{N-1,N}\}$$

and the matrix M as shown at the top of this paper.

Then, (A14) and (A15) are equivalent to

$$M\Im(\boldsymbol{\xi}_k) = 0, \ k = 1, \cdots, m \tag{A16}$$

and

$$M\Lambda\Re(\boldsymbol{\xi}_k) = 0, \ k = 1, \cdots, m \tag{A17}$$

respectively.

Since the system is transitionally non-degenerate, both M and Λ are nonsingular square matrices of order $[N(N-1)]/2$. Thus, (A16) and (A17) imply

$$\boldsymbol{\xi}_k = 0, \ k = 1, \cdots, m \tag{A18}$$

that is,

$$(H_k)_{jl}(p_l - p_j)\rho_{lj}(0) = 0, \ j, l = 1, \cdots, N, \ j < l \tag{A19}$$

Equation (A19) is the condition satisfied by the element $\rho_{lj}(0)$ of any state $\rho(0)$ in the largest invariant set E.

Thus, we complete the proof of Theorem 1. □

References

1 Shi S, Rabitz H. Quantum mechanical optimal control of physical observables in microsystems. *The Journal of Chemical Physics*, 1990, **92**(1): 364−376

2 Werschnik J, Gross E K U. Quantum optimal control theory. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 2007, **40**(18): 175−211

3 Shapiro E A, Milner V, Menzel-Jones C, Shapiro M. Piecewise adiabatic passage with a series of femtosecond pulses. *Physical Review Letters*, 2007, **99**(3): 2−5

4 Schirmer S G, Greentree A D, Ramakrishna V, Rabitz H. Constructive control of quantum systems using factorization of unitary operators. *Journal of Physics A: Mathematical and General*, 2002, **35**(39): 8315−8339

5 Grivopoulos S, Bamieh B. Lyapunov-based control of quantum systems. In: Proceedings of the 42nd IEEE Conference on Decision and Control. Maui, USA: IEEE, 2003. 434−438

6 Sugawara M. General formulation of locally designed coherent control theory for quantum system. *The Journal of Chemical Physics*, 2003, **118**(15): 6784−6800

7 Mirrahimi M, Rouchon P, Turinici G. Lyapunov control of bilinear Schroinger equations. *Automatica*, 2005, **41**(11): 1987−1994

8 Mirrahimi M, Turinici G, Rouchon P. Reference trajectory tracking for locally designed coherent quantum controls. *The Journal of Physical Chemistry A*, 2005, **109**(11): 2631−2637

9 Beauchard K, Coron J M, Mirrahimi M, Rouchon P. Implicit Lyapunov control of finite dimensional Schrodinger equations. *Systems and Control Letters*, 2007, **56**(5): 388−395

10 Cong Shuang, Kuang Sen. Quantum control strategy based on state distance. *Acta Automatica Sinica*, 2007, **33**(1): 28−31

11 Kuang S, Cong S. Lyapunov control methods of closed quantum systems. *Automatica*, 2008, **44**(1): 98−108

12 Wang X, Schirmer S G. Analysis of Lyapunov method for control of quantum states [Online], available: http://arxiv.org/abs/0901.4515, October 1, 2009

13 Wang X, Schirmer S G. Analysis of effectiveness of Lyapunov control for non-generic quantum states [Online], available: http://arxiv.org/abs/0901.4522, October 1, 2009

14 Altafini C. Feedback stabilization of isospectral control systems on complex flag manifolds: application to quantum ensembles. *IEEE Transactions on Automatic Control*, 2007, **52**(11): 2019−2028

15 Zhu W, Rabitz H. Quantum control design via adaptive tracking. *The Journal of Chemical Physics*, 2003, **119**(7): 3619−3625

16 Sugawara M, Tamaki M, Yabushita S. A new control scheme of multilevel quantum system based on effective decomposition by intense CW lasers. *The Journal of Physical Chemistry A*, 2007, **111**(38): 9446−9453

17 D′Alessandro D. *Introduction to Quantum Control and Dynamics*. Florida: CRC Press, 2007. 205−216

18 Schirmer S G, Zhang T, Leahy J V. Orbits of quantum states and geometry of Bloch vectors for *N*-level systems. *Journal of Physics A: Mathematical and General*, 2004, **37**(4): 1389−1402

19 Kimura G. The Bloch vector for *N*-level systems. *Physics Letters A*, 2003, **314**(5-6): 339−349

KUANG Sen Postdoctor at University of Science and Technology of China. He received his B. S. degree in industrial automation and M. S. degree in control theory and control engineering from Henan Polytechnic University in 1999 and 2003, respectively. And he received his Ph. D. degree in control theory and control engineering from University of Science and Technology of China in 2007. His research interest covers quantum feedback control and its state estimation, and quantum optimal control. Corresponding author of this paper. E-mail: skuang@ustc.edu.cn

CONG Shuang Professor in the Department of Automation, University of Science and Technology of China. She received her B. S. degree in automatic control from Beijing University of Aeronautics and Astronautics in 1982, and her Ph. D. degree in system engineering from University of Rome "La Sapienza", Rome, Italy in 1995, respectively. Her research interest covers advanced control strategies for motion control, fuzzy logic control, neural networks design and applications, robotic coordination control, and quantum system control. E-mail: scong@ustc.edu.cn