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Ensemble quantum control with a scalar input

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Abstract— In this article, we discuss how a three-level closed quantum system with dispersed parameters can be steered between eigenstates via a scalar control. The technique exploits a dynamical decoupling of the control based on the rotating wave approximation, which works under suitable conditions on the spectral gaps of the system and on the bounds on the parameter dispersion. We test numerically the sharpness of the conditions on several examples.

I. Introduction

Let us consider a continuum of three-level system described by the Schrödinger equation

$$i\dot{\psi}(t) = (H(\alpha) + \omega(t)H_c(\delta))\psi(t), \tag{1}$$

where $\omega(\cdot)$ is a real-valued control. Here the Hamiltonian $H(\alpha)$ is determined by an unknown parameter α taking values in a closed and connected subdomain $\mathcal{D}$ of $\mathbb{R}^m$ ($m \geq 1$). We assume that $H(\alpha)$ has the structure

$$H(\alpha) = \begin{pmatrix} \lambda_1(\alpha) & 0 & 0 \\ 0 & \lambda_2(\alpha) & 0 \\ 0 & 0 & \lambda_3(\alpha) \end{pmatrix}$$

where $\lambda_1(\cdot), \lambda_2(\cdot)$ and $\lambda_3(\cdot)$ are all continuous functions from $\mathcal{D}$ to $\mathbb{R}$. The matrix $H_c(\delta)$ is self-adjoint and describes the control coupling between eigenstates of the system

$$H(\delta) = \begin{pmatrix} 0 & \delta_{12} & \delta_{13} \\ \delta_{12} & 0 & \delta_{23} \\ \delta_{13} & \delta_{23} & 0 \end{pmatrix},$$

where δ_{jk} belongs to some closed interval $\mathcal{I}_{jk} = [\delta^0_{jk}, \delta^1_{jk}]$ in $\mathbb{R}$.

Previous results on the ensemble control problem deal mainly with the controllability of a two-level system with an unknown dispersion in its frequency and an unknown strength of coupling by its control. The controlled Hamiltonian in that case is given by

$$H(t) = \begin{pmatrix} E+\alpha & \omega(t) \\ \bar{\omega}(t) & -E-\alpha \end{pmatrix}. \tag{2}$$

In [1], [2], and [3], the authors considered system (2) steered by bounded complex controls. By using Lie algebra and adiabatic following arguments, a uniform control can be constructed to steer the system from a common initial state to an arbitrary set of target states continuously parameterized by (α, δ). An extension of these results to the case of real bounded controls has been obtained in [4]. Let us also mention [5], [6], [7] for some further results on ensemble control of quantum systems by adiabatic motion and [8], [9] for related ensemble stabilization problems for two-level quantum systems.

In this work we extend approaches previously proposed in [4] to three-level systems and underline the necessity of additional conditions due to the presence of a third state. In such a constructive approach, a rotating wave approximation and an adiabatic approximation are employed in cascade to realize a population inversion within the system. High-order averaging results have to be applied in the rotating wave and adiabatic steps, otherwise the fidelity of their cascade can fail to converge to 1, because of the competing time scales of these two approximations. The high-order averaging results require suitable conditions on the resonance frequencies that allow to validate the algorithm.

II. Results

Population inversion is a fundamental operation in many physical applications (see [10]) and is crucial for estabishing more general ensemble controllability (see [2], [4]). To realize a population inversion between the first and the second eigenstates, we will consider a time scale ϵ_1 for the rotating wave approximation and another time scale ϵ_2 for the adiabatic following. The control law in our algorithm will be a chirped pulse of the type

$$\omega_{\epsilon_1,\epsilon_2}(t) = 2\epsilon_1 u(\epsilon_1\epsilon_2 t)\cos\left(\phi_{\epsilon_1,\epsilon_2}(t)\right), \tag{3}$$

where

$$\phi_{\epsilon_1,\epsilon_2}(t) = \int_0^t f(\epsilon_1\epsilon_2\tau)\mathrm{d}\tau,$$

and $u, f : [0,T] \to \mathbb{R}$ are functions to be chosen. The goal is to induce an approximate transition from the initial state e_1 to a state of the form $\exp(i\theta_1)\mathrm{e}_2$, and similarly from e_2 to $\exp(i\theta_2)\mathrm{e}_1$. This is done for $\epsilon_1, \epsilon_2 \to 0$ in time $T/(\epsilon_1\epsilon_2)$.

In the following, we will denote by $\{\mathrm{e}_1, \mathrm{e}_2, \mathrm{e}_3\}$ the canonical basis of $\mathbb{R}^3$ and $\{\mathrm{e}_{jk}\}_{(j,k)\in\{1,2,3\}^2}$ the canonical basis of 3×3 real matrices.

Remark 1: It should be noticed that the technique that we propose here does not permit to realize a STIRAP [11], [12], [13] transfer in a three-level system with one scalar control only .[1] This looks to be a very hard task.

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[1] Namely, a transition from the state 1 to the state 3, with minimal population of the state 2, by using a control containing two frequencies: first the resonance frequency between state 2 and 3 and then the resonance frequency between state 1 and 2 (the famous counter-intuitive strategy).

Theorem 2: Let us assume that δ_{12} is in a closed interval $\mathcal{I}_{12} = [\delta_{12}^0, \delta_{12}^1]$ such that $0 \notin \mathcal{I}_{12}$ and that for all $\alpha \in \mathcal{D}$, $\lambda_2(\alpha) - \lambda_1(\alpha) > 0$, $|\lambda_1(\alpha) - \lambda_3(\alpha)| > 0$ and $|\lambda_2(\alpha) - \lambda_3(\alpha)| > 0$. Assume that there exists $0 < v_0 < v_1$ such that

1) For all $\alpha \in \mathcal{D}$, $\lambda_2(\alpha) - \lambda_1(\alpha) \in (v_0, v_1)$ and, for $(j,k) = (1,3)$ or $(2,3)$, we have that $\forall \alpha \in \mathcal{D}$, $|\lambda_j(\alpha) - \lambda_k(\alpha)| \notin [v_0, v_1]$.
2) For all $1 \le j < k \le 3$, we have that $\forall \alpha \in \mathcal{D}$, $|\lambda_k(\alpha) - \lambda_j(\alpha)| \notin [2v_0, 2v_1]$.

Then we can fix $T > 0$ and take $u, f \in \mathcal{C}^2([0,T], \mathbb{R})$ such that

i) $(u(0), f(0)) = (0, v_0)$ and $(u(T), f(T)) = (0, v_1)$;
ii) $\forall s \in (0,T), u(s) > 0, \dot{f}(s) > 0$.

Denote by $\psi_{\epsilon_1,\epsilon_2}$ the solution of (1) with initial condition $\psi_{\epsilon_i,\epsilon_2}(0) = \mathrm{e}_1$ and the control law $\omega_{\epsilon_1,\epsilon_2}$ as in (3). Then there exist $C > 0$ and $\eta > 0$ such that for every $\alpha \in \mathcal{D}$ and every $(\epsilon_1, \epsilon_2) \in (0, \eta)^2$,

$$\left\| \psi_{\epsilon_1,\epsilon_2}\left(\frac{T}{\epsilon_1\epsilon_2}\right) - \exp(i\theta)\mathrm{e}_2 \right\| < C \max\left(\frac{\epsilon_1^2}{\epsilon_2}, \frac{\epsilon_2}{\epsilon_1}\right) \quad (4)$$

for some $\theta \in \mathbb{R}$. The same result holds for the initial state e_2 and the final state $\exp(i\beta)\mathrm{e}_1$ for some $\beta \in \mathbb{R}$.

Remark 3: For a system with no dispersion in its frequencies, the non-resonance of gaps between eigenvalues is known to be crucial to establish controllability (see, e.g., [14]). Our theorem provides a generalization of this condition to the ensemble control problem.

Remark 4: If we choose $\epsilon_2 = \epsilon_1^{3/2}$, we have

$$\min_{\theta \in [0,2\pi]} \left\| \psi_{\epsilon_1,\epsilon_2}\left(\frac{T}{\epsilon_1\epsilon_2}\right) - \exp(i\theta)\mathrm{e}_2 \right\| < C\epsilon_1^{1/2},$$

that is, the final state is arbitrarily close to the eigenstate e_2, up to a phase, as ϵ_1 goes to zero.

Remark 5: Here we fix $T = 1$ and we give a simple example of $u(\cdot)$ and $f(\cdot)$ satisfying the conditions of Theorem 2:

$$u(s) = \sin(\pi s), \quad f(s) = \frac{v_0 + v_1}{2} + \frac{v_0 - v_1}{2}\cos(\pi s).$$

Thus the control law is given by

$$\begin{aligned} \omega_{\epsilon_1,\epsilon_2}(t) = 2\epsilon_1 \sin(\epsilon_1\epsilon_2\pi t)\cos\Big(&\frac{v_1+v_0}{2}t \\ &+ \frac{v_0 - v_1}{2\epsilon_1\epsilon_2\pi}\sin(\epsilon_1\epsilon_2\pi t)\Big), \quad t \in \left[0, \frac{1}{\epsilon_1\epsilon_2}\right]. \end{aligned} \quad (5)$$

III. Proof of the Theorem

For $E \in \mathbb{R}$ and $1 \le j \le k \le 3$, let us define

$$A_{jk}(E) = \begin{cases} \exp(iE)\mathrm{e}_{jk} + \exp(-iE)\mathrm{e}_{kj} & \text{if } j < k, \\ \cos(E)\mathrm{e}_{jj} & \text{if } j = k, \end{cases}$$

$$B_{jk}(E) = \begin{cases} i\exp(iE)\mathrm{e}_{jk} - i\exp(-iE)\mathrm{e}_{kj} & \text{if } j < k, \\ -\sin(E)\mathrm{e}_{jj} & \text{if } j = k. \end{cases}$$

Let us recast (1) in the interaction frame

$$\psi(t) = \exp(-itH(\alpha))\psi_I(t).$$

Notice that

$$i\frac{\mathrm{d}}{\mathrm{d}t}\psi_I = H_I(t)\psi(t), \quad (6)$$

where

$$\begin{aligned} H_I(t) &= -H(\alpha) + \exp(itH(\alpha))H(t)\exp(-itH(\alpha)) \\ &= \omega_{\epsilon_1,\epsilon_2}(t)\Big(\delta_{12}A_{12}\big((\lambda_1 - \lambda_2)t\big) + \delta_{13}A_{13}\big((\lambda_1 - \lambda_3)t\big) \\ &+ \delta_{23}A_{23}\big((\lambda_2 - \lambda_3)t\big)\Big) \end{aligned}$$

For $1 \le j \le k \le 3$ and $\sigma \in \{\pm 1, \pm 2\}$, let us define:

$$\begin{aligned} \phi_{jk}^\sigma(t) &= (\lambda_j - \lambda_k)t + \sigma\phi_{\epsilon_1,\epsilon_2}(t), \\ f_{jk}^\sigma(t) &= \frac{\mathrm{d}}{\mathrm{d}t}\phi_{jk}^\sigma(t) = \lambda_j - \lambda_k + \sigma f(\epsilon_1\epsilon_2 t). \end{aligned} \quad (7)$$

Then, with the control $\omega_{\epsilon_1,\epsilon_2}(\cdot)$ given in Equation (3), we have

$$\begin{aligned} H_I(t) = \epsilon_1 u(\epsilon_1\epsilon_2 t)\Big(&\delta_{12}A_{12}\big(\phi_{12}^1(t)\big) + \delta_{12}A_{12}\big(\phi_{12}^{-1}(t)\big) \\ &+ \delta_{13}A_{13}\big(\phi_{13}^1(t)\big) + \delta_{13}A_{13}\big(\phi_{13}^{-1}(t)\big) \\ &+ \delta_{23}A_{23}\big(\phi_{23}^1(t)\big) + \delta_{23}A_{23}\big(\phi_{23}^{-1}(t)\big)\Big). \end{aligned}$$

Remark 6: Consider the change of variables

$$\psi(t) = \exp(ix(t))\tilde{\psi}(t), \quad (8)$$

where $x(\cdot)$ is a smooth curve in the space of $n \times n$ Hermitian matrices and $\psi(\cdot)$ is the solution of the Schrödinger equation

$$i\frac{\mathrm{d}}{\mathrm{d}t}\psi(t) = H(t)\psi(t).$$

Then it can be easily verified that $\tilde{\psi}(\cdot)$ is the solution of

$$i\frac{\mathrm{d}}{\mathrm{d}t}\tilde{\psi}(t) = \tilde{H}(t)\tilde{\psi}(t), \quad \tilde{\psi}(0) = \exp(-ix(0))\psi(0),$$

where $\tilde{H}(\cdot)$ is given by

$$\tilde{H}(t) = \mathrm{Ad}_{\exp(-ix(t))}H(t) + \mathrm{d}\exp(ix(t))\frac{\mathrm{d}}{\mathrm{d}t}x(t). \quad (9)$$

Here, for an $n \times n$ Hermitian matrix h, $\mathrm{d}\exp(ix(t))$ and $\mathrm{Ad}_{\exp(-ix(t))}$ are the automorphisms defined by

$$\begin{aligned} \mathrm{d}\exp(ix(t))h &= \exp(-ix(t))\Big((D\exp)(ix(t))h\Big), \\ \mathrm{Ad}_{\exp(-ix(t))}h &= \exp(-ix(t))h\exp(ix(t)), \end{aligned}$$

where $D\exp(ix(t))$ denotes the differential of the exponential mapping at $ix(t)$.

Definition 7: We call R a (ϵ_1, ϵ_2)-parameterized function if for every $\epsilon_1, \epsilon_2 > 0$, $R_{\epsilon_1,\epsilon_2}$ is a real valued function defined on $\left[0, \frac{1}{\epsilon_1\epsilon_2}\right]$. Given an (ϵ_1, ϵ_2)-parameterized function R and $g : \mathbb{R}_+^2 \to \mathbb{R}_+$, we say that $R_{\epsilon_1,\epsilon_2} = \mathcal{O}\big(g(\epsilon_1, \epsilon_2)\big)$ if there exist $\delta, C > 0$ such that for every $(\epsilon_1, \epsilon_2) \in (0, \delta)^2$ and $t \in \left[0, \frac{1}{\epsilon_1\epsilon_2}\right]$, we have $|R_{\epsilon_1,\epsilon_2}(t)| \le Cg(\epsilon_1, \epsilon_2)$

a) First-order elimination: Let us first define the sets of indices

$$\mathcal{I} = \{(j,k,\sigma) \mid 1 \le j < k \le 3, \sigma = \pm 1\},$$
$$\mathcal{I}' = \mathcal{I} \setminus \{(1,2,1)\}.$$

Assume that hypothesis 1 (first-order condition) of Theorem 2 are satisfied. Then, for every $(j,k,\sigma) \in \mathcal{I}'$,

$$f_{jk}^{\sigma}(t) = \lambda_j - \lambda_k + \sigma f(\epsilon_1\epsilon_2 t) \neq 0, \quad \forall t \in \left[0, \frac{1}{\epsilon_1\epsilon_2}\right],$$

where $f_{jk}^{\sigma}(\cdot)$ is introduced in Equation (7). Then we can apply a first change of variables to system (6) in the interaction frame

$$\psi_I(t) = \exp(i\epsilon_1 X_1(t))\hat{\psi}_1(t),$$

where

$$X_1(t) = \sum_{(j,k,\sigma)\in\mathcal{I}'} \delta_{jk} c_{jk}^{\sigma}(\epsilon_1\epsilon_2 t) B_{jk}\big(\phi_{jk}^{\sigma}(t)\big),$$

where for every $(j,k,\sigma) \in \mathcal{I}'$ and $s \in [0,1]$, we have

$$c_{jk}^{\sigma}(s) = \frac{u(s)}{f_{jk}^{\sigma}(s)}.$$

Notice that for every $(j,k,\sigma) \in \mathcal{I}'$,

$$\frac{\mathrm{d}}{\mathrm{d}t}\Big(c_{jk}(\epsilon_1\epsilon_2 t) B_{jk}\big(\phi_{jk}^{\sigma}(t)\big)\Big) = -A_{jk}\big(\phi_{jk}^{\sigma}(t)\big) + \mathcal{O}(\epsilon_1\epsilon_2).$$

where $\mathcal{O}(\cdot)$ is defined as in Definition 7. Then, by differentiating X_1, we obtain that

$$\frac{\mathrm{d}}{\mathrm{d}t}X_1(t) = -\sum_{(j,k,\sigma)\in\mathcal{I}'} \delta_{jk} u(\epsilon_1\epsilon_2 t) A_{jk}\big(\phi_{jk}^{\sigma}(t)\big) + \mathcal{O}(\epsilon_1\epsilon_2).$$

If we use $x(t) = \epsilon_1 X_1(t)$ in (9), then by Baker-Hausdorff Formula and by Theorem 4.5 in [15], we deduce that the dynamics of $\hat{\psi}_1$ are characterized by the Hamiltonian

$$\begin{aligned}\hat{H}_1(t) =& H_I(t) + \epsilon_1 \frac{\mathrm{d}}{\mathrm{d}t}X_1(t) \\ &- i\epsilon_1 \left[X_1(t), H_I(t) + \epsilon_1 \frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}X_1(t)\right] + \mathcal{O}(\epsilon_1^3).\end{aligned} \tag{10}$$

Notice that

$$\begin{aligned}H_I(t) + \epsilon_1 \frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}X_1(t) &= \epsilon_1 u(\epsilon_1\epsilon_2 t)\delta_{12} A_{12}\big(\phi_{12}^{1}(t)\big) \\ &+ \sum_{(j,k,\sigma)\in\mathcal{I}'} \frac{1}{2}\epsilon_1 u(\epsilon_1\epsilon_2 t)\delta_{jk} A_{jk}(\phi_{jk}^{\sigma}(t)).\end{aligned}$$

It can be deduced that (10) will have the following structure:

$$\begin{aligned}&\hat{H}_1(t) = \epsilon_1 \delta_{12} u(\epsilon_1\epsilon_2 t) A_{12}\big(\phi_{12}^{1}(t)\big) \\ &+ \sum_{(j,k,\sigma)\in\mathcal{J}} \epsilon_1^2 h_{jk}^{\sigma}(\epsilon_1\epsilon_2 t) A_{jk}\big(\phi_{jk}^{\sigma}(t)\big) + \mathcal{O}(\epsilon_1^3 + \epsilon_1^2\epsilon_1).\end{aligned} \tag{11}$$

where

$$\mathcal{J} = \{(j,k,\sigma) \mid 1 \le j \le k \le 3, \sigma \in \{-2,0,2\}\}.$$

Here one should notice that, since $u(0) = u(1) = 0$, we have $h_{jk}^{\sigma}(0) = h_{jk}^{\sigma}(1) = 0$ and that h_{jk}^{σ} is independent of (ϵ_1, ϵ_2).

b) Second-order elimination: Set

$$\mathcal{J}' = \mathcal{J} \setminus \{(j,j,0) \mid 1 \le j \le 3\}.$$

Hypothesis 2 (second-order condition) of Theorem 2 implies that, for every $(j,k,\sigma) \in \mathcal{J}'$,

$$f_{jk}^{\sigma}(t) = \lambda_j - \lambda_k + \sigma f(\epsilon_1\epsilon_2 t) \neq 0, \quad \forall t \in \left[0, \frac{1}{\epsilon_1\epsilon_2}\right].$$

Let us introduce a second change of variables

$$\hat{\psi}_1(t) = \exp(i\epsilon_1^2 X_2(t))\hat{\psi}_2(t),$$

where

$$X_2(t) = \sum_{(j,k,\sigma)\in\mathcal{J}'} \frac{h_{jk}^{\sigma}(\epsilon_1\epsilon_2 t)}{f_{jk}^{\sigma}(\epsilon_1\epsilon_2 t)} B_{jk}\big(\phi_{jk}^{\sigma}(t)\big). \tag{12}$$

Notice that

$$\frac{\mathrm{d}}{\mathrm{d}t}X_2(t) = -\sum_{(j,k,\sigma)\in\mathcal{J}'} h_{jk}^{\sigma}(\epsilon_1\epsilon_2 t) A_{jk}\big(\phi_{jk}^{\sigma}(t)\big) + \mathcal{O}(\epsilon_1\epsilon_2).$$

Then, applying a method similar to that used for first-order elimination, we deduce that the dynamics of $\hat{\psi}_2(t)$ is characterized by the Hamiltonian

$$\begin{aligned}&\hat{H}_2(t) = \hat{H}_1(t) + \epsilon_1^2 \frac{\mathrm{d}}{\mathrm{d}t}X_2(t) + \mathcal{O}(\epsilon_1^3) \\ &= \epsilon_1 \delta_{12} u(\epsilon_1\epsilon_2 t) A_{12}\big(\phi_{12}^{1}(t)\big) \\ &+ \sum_{j=1}^{3} \epsilon_1^2 h_{jj}^{0}(\epsilon_1\epsilon_2 t) A_{jj}(0) + \mathcal{O}(\epsilon_1^3 + \epsilon_1^2\epsilon_2).\end{aligned} \tag{13}$$

Remark 8: Since $u(0) = u(1) = 0$ and $h_{jk}^{\sigma}(0) = h_{jk}^{\sigma}(1) = 0$ for every $(j,k,\sigma) \in \mathcal{J}$, we can deduce that $X_1(0) = X_1\left(\frac{1}{\epsilon_1\epsilon_2}\right) = 0$ and $X_2(0) = X_2\left(\frac{1}{\epsilon_1\epsilon_2}\right) = 0$. Hence, $\psi_I(0) = \hat{\psi}_2(0)$ and $\psi_I\left(\frac{1}{\epsilon_1\epsilon_2}\right) = \hat{\psi}_2\left(\frac{1}{\epsilon_2\epsilon_2}\right)$.

c) Rotating wave approximation: Let us introduce the following truncation of $\hat{H}_2(t)$

$$\begin{aligned}H_{\mathrm{RWA}}(t) &= \epsilon_1 \delta_{12} u(\epsilon_1\epsilon_2 t) A_{12}\big(\phi_{12}^{1}(t)\big) \\ &+ \sum_{j=1}^{3} \epsilon_1^2 h_{jj}^{0}(\epsilon_1\epsilon_2 t) \mathrm{e}_{jj}.\end{aligned} \tag{14}$$

We denote by ψ_{RWA} the solution of the system

$$i\frac{\mathrm{d}}{\mathrm{d}t}\psi_{\mathrm{RWA}}(t) = H_{\mathrm{RWA}}(t)\psi_{\mathrm{RWA}}(t), \quad \psi_{\mathrm{RWA}}(0) = \psi_I(0),$$

where ψ_I is the solution of system (6).

Lemma 9: We have that

$$\left\|\psi_I\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \psi_{\mathrm{RWA}}\left(\frac{1}{\epsilon_1\epsilon_2}\right)\right\| = \mathcal{O}\left(\frac{\epsilon_1^2}{\epsilon_2} + \epsilon_1\right).$$

Proof: Introduce the $SU(3)$-valued functions $U(\cdot), W(\cdot)$ solutions of

$$i\frac{\mathrm{d}}{\mathrm{d}t}U(t) = \hat{H}_2(t)U(t), \quad U(0) = \mathbb{I}_3,$$
$$i\frac{\mathrm{d}}{\mathrm{d}t}W(t) = H_{\mathrm{RWA}}(t)W(t), \quad W(0) = \mathbb{I}_3.$$

It is evident that $\hat{\psi}_2(t) = U(t)\psi_I(0)$ and $\psi_{\mathrm{RWA}}(t) = W(t)\psi_I(0)$ for every $t \in \left[0, \frac{1}{\epsilon_1\epsilon_2}\right]$. By differentiating $W^\dagger(t)U(t)$ with respect to t, we obtain that

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(W^\dagger(t)U(t)\right) = -iW^\dagger(t)\left(\hat{H}_2(t) - H_{\mathrm{RWA}}(t)\right)U(t).$$

Since $\hat{H}_2(t) - H_{\mathrm{RWA}}(t) = \mathcal{O}(\epsilon_1^3 + \epsilon_1^2\epsilon_2)$ in sense of $\mathcal{O}(\cdot)$ defined in Definition 7, then

$$\begin{aligned}
&\left\|W^\dagger\left(\frac{1}{\epsilon_1\epsilon_2}\right)U\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \mathbb{I}_3\right\| \\
&\leq \int_0^{\frac{1}{\epsilon_1\epsilon_2}} \left\|\frac{\mathrm{d}}{\mathrm{d}t}\left(W^\dagger(t)U(t)\right)\right\| \mathrm{d}t \\
&= \int_0^{\frac{1}{\epsilon_1\epsilon_2}} \left\|W^\dagger(t)\left(\hat{H}_2(t) - H_{\mathrm{RWA}}(t)\right)U(t)\right\| \mathrm{d}t \\
&= \int_0^{\frac{1}{\epsilon_1\epsilon_2}} \left\|\hat{H}_2(t) - H_{\mathrm{RWA}}(t)\right\| \mathrm{d}t = \mathcal{O}\left(\frac{\epsilon_1^2}{\epsilon_2} + \epsilon_1\right).
\end{aligned}$$

Since $\psi_I\left(\frac{1}{\epsilon_1\epsilon_2}\right) = \hat{\psi}_2\left(\frac{1}{\epsilon_1\epsilon_2}\right)$ (see Remark 8), we obtain that

$$\begin{aligned}
&\left\|\psi_I\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \psi_{\mathrm{RWA}}\left(\frac{1}{\epsilon_1\epsilon_2}\right)\right\| \\
&= \left\|\hat{\psi}_2\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \psi_{\mathrm{RWA}}\left(\frac{1}{\epsilon_1\epsilon_2}\right)\right\| \\
&= \left\|\left(U\left(\frac{1}{\epsilon_1\epsilon_2}\right) - W\left(\frac{1}{\epsilon_1\epsilon_2}\right)\right)\psi_I(0)\right\| \\
&= \left\|W\left(\frac{1}{\epsilon_1\epsilon_2}\right)\left(W^\dagger\left(\frac{1}{\epsilon_1\epsilon_2}\right)U\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \mathbb{I}_3\right)\psi_I(0)\right\| \\
&\leq \left\|W^\dagger\left(\frac{1}{\epsilon_1\epsilon_2}\right)U\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \mathbb{I}_3\right\| = \mathcal{O}\left(\frac{\epsilon_1^2}{\epsilon_2} + \epsilon_1\right).
\end{aligned}$$

■

d) Adiabatic following: Let us introduce the unitary change of variables $\psi_{\mathrm{RWA}}(t) = U(t)\psi_{\mathrm{slow}}(t)$ with

$$U(t) = \begin{pmatrix} e^{i\lambda_1(\alpha)t} & 0 & 0 \\ 0 & e^{i(\lambda_2(\alpha)t - \int_0^t f(\epsilon_1\epsilon_2\tau)\mathrm{d}\tau)} & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (15)$$

Then $i\frac{\mathrm{d}}{\mathrm{d}t}\psi_{\mathrm{slow}}(t) = H_{\mathrm{slow}}(t)\psi_{\mathrm{slow}}(t)$, where

$$\begin{aligned}
H_{\mathrm{slow}}(t) = &\begin{pmatrix} \lambda_1(\alpha) & \epsilon_1\delta_{12}u(\epsilon_1\epsilon_2 t) & 0 \\ \epsilon_1\delta_{12}u(\epsilon_1\epsilon_2 t) & \lambda_2(\alpha) - f(\epsilon_1\epsilon_2 t) & 0 \\ 0 & 0 & 0 \end{pmatrix} \\
&+\epsilon_1^2\begin{pmatrix} h_{11}^0(\epsilon_1\epsilon_2 t) & 0 & 0 \\ 0 & h_{22}^0(\epsilon_1\epsilon_2 t) & 0 \\ 0 & 0 & h_{33}^0(\epsilon_1\epsilon_2 t) \end{pmatrix}.
\end{aligned} \quad (16)$$

It is evident that the dynamics in the two-dimensional space $\mathrm{span}(\mathrm{e}_1, \mathrm{e}_2)$ is decoupled from the rest of the system. Let us define the decoupled Hamiltonian in $\mathrm{span}(\mathrm{e}_1, \mathrm{e}_2)$

$$\begin{aligned}
H_{\mathrm{slow}}^d(t) = &\begin{pmatrix} \lambda_1(\alpha) & \epsilon_1\delta_{12}u(\epsilon_1\epsilon_2 t) \\ \epsilon_1\delta_{12}u(\epsilon_1\epsilon_2 t) & \lambda_2(\alpha) - f(\epsilon_1\epsilon_2 t) \end{pmatrix} \\
&+ \epsilon_1^2\begin{pmatrix} h_{11}^0(\epsilon_1\epsilon_2 t) & 0 \\ 0 & h_{22}^0(\epsilon_1\epsilon_2 t) \end{pmatrix}.
\end{aligned}$$

If $\psi_{\mathrm{slow}}(0) \in \mathrm{span}(\mathrm{e}_1, \mathrm{e}_2)$, the solution ψ_{slow}^d of

$$i\frac{\mathrm{d}}{\mathrm{d}t}\psi_{\mathrm{slow}}^d(t) = H_{\mathrm{slow}}^d(t)\psi_{\mathrm{slow}}^d(t), \quad \psi_{\mathrm{slow}}^d(0) = \psi_{\mathrm{slow}}(0), \quad (17)$$

satisfies $\psi_{\mathrm{slow}}^d(t) = \psi_{\mathrm{slow}}(t)$ for every $t \in \left[0, \frac{1}{\epsilon_1\epsilon_2}\right]$. Then if δ_{12} is in a closed interval $\mathcal{I}_{12} = [\delta_{12}^0, \delta_{12}^1]$ such that $0 \notin \mathcal{I}_{12}$, we can apply Lemma 30 in [4] to the decoupled two-level system and obtain that, for the control $\omega(\cdot)$ given in Theorem 2, there exists $C' > 0$ such that for $\psi_{\mathrm{slow}}(0) = \mathrm{e}_1$

$$\min_{\theta\in[0,2\pi]}\left\|\psi_{\mathrm{slow}}\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \exp(i\theta)\mathrm{e}_2\right\| \leq C'\frac{\epsilon_2}{\epsilon_1}.$$

Then by the change of variables introduced in Equation (15) and the estimation given in Lemma 9, we deduce that, in the interaction frame, there exists $C > 0$ such that

$$\min_{\theta\in[0,2\pi]}\left\|\psi_I\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \exp(i\theta)\mathrm{e}_2\right\| \leq C\max\left(\frac{\epsilon_1^2}{\epsilon_2}, \frac{\epsilon_2}{\epsilon_1}\right).$$

It follows that

$$\min_{\theta\in[0,2\pi]}\left\|\psi_{\epsilon_1,\epsilon_2}\left(\frac{1}{\epsilon_1\epsilon_2}\right) - \exp(i\theta)\mathrm{e}_2\right\| < C\max\left(\frac{\epsilon_1^2}{\epsilon_2}, \frac{\epsilon_2}{\epsilon_1}\right).$$

Notice that all the reasoning above holds for the initial state e_2 and the final state of the form $\exp(i\beta)\mathrm{e}_1$. This concludes the proof of Theorem 2.

IV. Example

Consider a system as in (1) with drift and control Hamiltonians

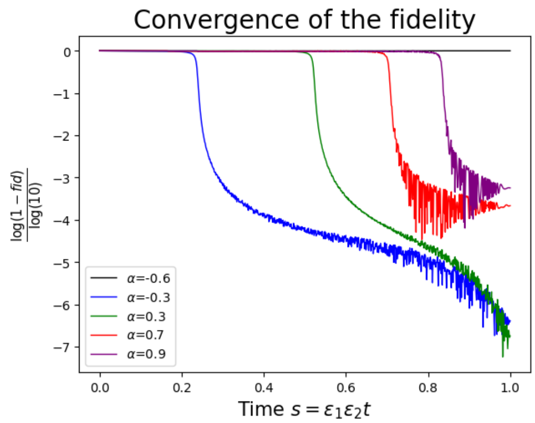
$$H(\alpha) = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1.5+\alpha & 0 \\ 0 & 0 & 2+\beta \end{pmatrix}, \quad H_c = \begin{pmatrix} 0 & 1 & 5 \\ 1 & 0 & 5 \\ 5 & 5 & 0 \end{pmatrix},$$

First, let us fix $\beta = 6$ and $\psi_{\epsilon_1,\epsilon_2}(0) = \mathrm{e}_1$. In order to realize a population inversion between the first and the second eigenstates, we fix $T = 1$ and use the control law given in (5) with $v_0 = 1$, $v_1 = 2.5$. To test the sharpness of conditions, we will use five different values of α: $\alpha = -0.6$, $\alpha = -0.3$, $\alpha = 0.3$, $\alpha = 0.7$, $\alpha = 0.9$ and two pairs of time scales: $(\epsilon_1, \epsilon_2) = (10^{-2}, 10^{-7/2})$ and $(\epsilon_1, \epsilon_2) = (10^{-3/2}, 10^{-3})$. The fidelity at $s \in [0, 1]$ is defined as

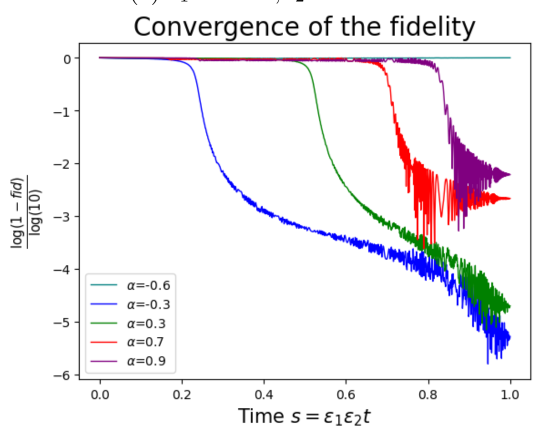
$$\mathrm{fid}(s) = 1 - \min_{\theta\in[0,2\pi]}\left\|\psi_{\epsilon_1,\epsilon_2}\left(\frac{s}{\epsilon_1\epsilon_2}\right) - e^{i\theta}\mathrm{e}_2\right\|. \quad (18)$$

To better illustrate the convergence of fidelity to 1, the vertical axis of the figures will be scaled logarithmically, specifically $\log(1 - \mathrm{fid}(s))/\log 10$. For every $\alpha \in \{-0.6, -0.3, 0.3, 0.7, 0.9\}$, $|\lambda_3(\alpha, \beta) - \lambda_1(\alpha, \beta)|$ and $|\lambda_3(\alpha, \beta) - \lambda_2(\alpha, \beta)|$ are not in $[v_0, v_1] \cup [2v_0, 2v_1]$. When $\alpha = -0.6$, $\lambda_2(\alpha, \beta) - \lambda_1(\alpha, \beta) \notin [v_0, v_1]$, violating Hypothesis 1 of Theorem 2. When $\alpha \in \{-0.3, 0.3\}$, both Hypotheses 1 and 2 of Theorem 2 are satisfied. When $\alpha \in \{0.7, 0.9\}$, $\lambda_2(\alpha) - \lambda_1(\alpha) \in [2v_0, 2v_1]$ and Hypothesis 2 is violated. See results in Figures 1a and 1b. Notice that the convergence seems to be achieved even though

$(\epsilon_1, \epsilon_2) = (10^{-3/2}, 10^{-3})$ does not guarantee a small error in Equation (4) (see also Remark 4). This observation suggests the possibility of refining the error estimation using higher-order averaging.



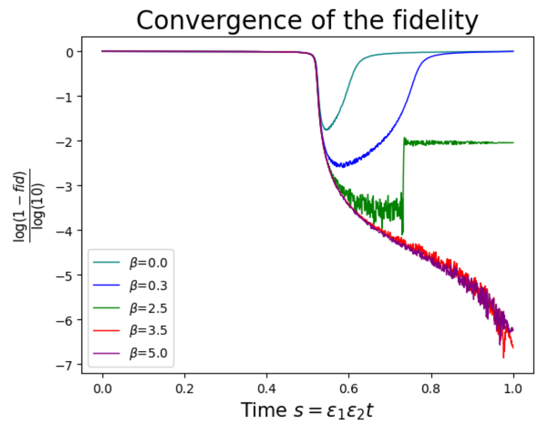
(a) $\epsilon_1 = 10^{-2}, \epsilon_2 = 10^{-7/2}$



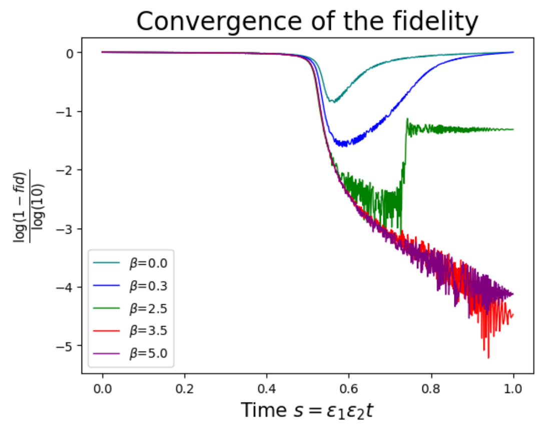
(b) $\epsilon_1 = 10^{-3/2}, \epsilon_2 = 10^{-3}$,

Fig. 1: Simulations for $\beta = 6$, $\alpha \in \{-0.6, -0.3, 0.3, 0.7, 0.9\}$: Convergence holds only when the first-order condition (Hypothesis 1) is satisfied and the best convergence is obtained when both the first-order condition and the second-order condition (Hypothesis 1 and Hypothesis 2) are satisfied.

Then let us fix $\alpha = 0.3$ and $\psi_{\epsilon_1,\epsilon_2}(0) = \mathrm{e}_2$. With the same control signal as before, we will test the fidelity of population inversion between the first and the second eigenstates for different values of β: $\beta = 0$, $\beta = 0.3$, $\beta = 2.5$, $\beta = 3.5$, $\beta = 5.0$. We also consider two pairs of time scales: $(\epsilon_1, \epsilon_2) = (10^{-2}, 10^{-7/2})$ and $(\epsilon_1, \epsilon_2) = (10^{-3/2}, 10^{-3})$. The fidelity is computed as in Equation (18) with the target state e_1. Notice that, for every possible value of β, $\lambda_2(\alpha,\beta) - \lambda_1(\alpha,\beta) \in [v_0, v_1]$ and $|\lambda_2(\alpha,\beta) - \lambda_1(\alpha,\beta)| \notin [2v_0, 2v_1]$. When $\beta \in \{0.0, 0.3\}$, we have $\lambda_3(\alpha,\beta) - \lambda_1(\alpha,\beta) \in [v_0, v_1]$, violating Hypothesis 1. When $\beta = 2.5$, we have $\lambda_3(\alpha,\beta) - \lambda_1(\alpha.\beta) \in [2v_0, 2v_1]$, violating Hypothesis 2. When $\beta \in \{3.5, 5.0\}$, both hypotheses are satisfied. See results in Figures 2a and 2b.



(a) $\epsilon_1 = 10^{-2}, \epsilon_2 = 10^{-7/2}$



(b) $\epsilon_1 = 10^{-3/2}, \epsilon_2 = 10^{-3}$,

Fig. 2: Simulations for $\alpha = 0.3$, $\beta \in \{0.0, 0.3, 2.5, 3.5, 5.0\}$: Convergence holds when both first-order and second-order conditions are satisfied and is lost when the first-order condition is violated. Notice that for $\beta = 2.5$ an abrut variation of the fidelity (in the log scale) occurs when $\lambda_3(\alpha,\beta) - \lambda_2(\alpha,\beta) - 2f(s) = 0$, that is, at $s = \arccos(-2/3)/\pi \approx 0.7323$.

Finally, let us test if it is possible to realize a population inversion e_1 and e_3 by successive population inversions between $(\mathrm{e}_1, \mathrm{e}_2)$ and then between $(\mathrm{e}_2, \mathrm{e}_3)$. In order to realize a population inversion between $(\mathrm{e}_1, \mathrm{e}_2)$, let us fix $T = 1$, $\psi_{\epsilon_1,\epsilon_2}(0) = \mathrm{e}_1$. We use $v_0 = 1, v_1 = 2.5$ and the control law given in Equation (5) to construct a control law $\omega^1_{\epsilon_1,\epsilon_2}(\cdot)$ defined on $\left[0, \frac{1}{\epsilon_1\epsilon_2}\right]$. Notice that the hypotheses of Theorem 2 are verified. Then, in order to realize a population inversion between $(\mathrm{e}_2, \mathrm{e}_3)$, we can set $(\tilde{\mathrm{e}}_1, \tilde{\mathrm{e}}_2, \tilde{\mathrm{e}}_3) = (\mathrm{e}_2, \mathrm{e}_3, \mathrm{e}_1)$ and $(\tilde{\lambda}_1, \tilde{\lambda}_2, \tilde{\lambda}_3) = (\lambda_2, \lambda_3, \lambda_1)$ and apply Theorem 2. Let us use $v_0 = 5, v_1 = 7$ to construct the control law $\omega^2_{\epsilon_1,\epsilon_2}(\cdot)$. It can also be easily verified that the hypotheses of Theorem 2 are verified. A concatenation of these two control laws defined on $\left[0, \frac{2}{\epsilon_1\epsilon_2}\right]$ is given by

$$\omega_{\epsilon_1,\epsilon_2} : t \mapsto \begin{cases} \omega^1_{\epsilon_1\epsilon_2}(t) & \text{if } t < 1/(\epsilon_1\epsilon_2) \\ \omega^2_{\epsilon_1\epsilon_2}\big(t - 1/(\epsilon_1\epsilon_2)\big) & \text{if } t \geq 1/(\epsilon_1\epsilon_2) \end{cases}$$

The numerical result is given in Figure 3 with the time scale $(\epsilon_1, \epsilon_2) = (10^{-4/3}, 10^{-3})$, showing the efficacy of the proposed algorithm.

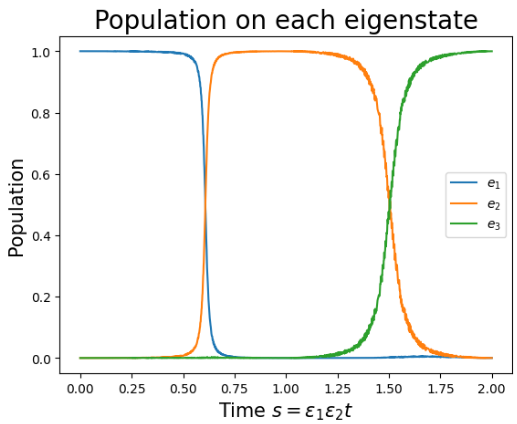


Fig. 3: Simulations for $\alpha = 0.5$, $\beta = 6$, $\epsilon_1 = 10^{-4/3}, \epsilon_2 = 10^{-3}$: Population inversions between $(\mathrm{e}_1, \mathrm{e}_2)$ and $(\mathrm{e}_2, \mathrm{e}_3)$ happen successively.

V. Conclusion

In this study, we introduced an algorithm capable of realizing population inversion between the two first eigenstates for a three-level systems. We underlined the importance of non-overlapping of some characteristic frequencies for this algorithm's validity. Future investigations could explore the possibility of proposing weaker conditions for convergence, generalizing this constructive method to n-level or even infinite-dimensional quantum systems and examining which further controllability results (i.e., population splitting) could be obtained through population inversion.

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References

[1] J.-S. Li and N. Khaneja, “Ensemble controllability of the Bloch equations,” in Proceedings of the 45th IEEE Conference on Decision and Control. IEEE, 2006, pp. 2483–2487.

[2] ——, “Ensemble control of Bloch equations,” IEEE Transactions on Automatic Control, vol. 54, no. 3, pp. 528–536, 2009.

[3] K. Beauchard, J.-M. Coron, and P. Rouchon, “Controllability issues for continuous-spectrum systems and ensemble controllability of Bloch equations,” Comm. Math. Phys., vol. 296, no. 2, pp. 525–557, 2010. [Online]. Available: https://doi.org/10.1007/s00220-010-1008-9

[4] R. Robin, N. Augier, U. Boscain, and M. Sigalotti, “Ensemble qubit controllability with a single control via adiabatic and rotating wave approximations,” Journal of Differential Equations, vol. 318, pp. 414–442, 2022.

[5] N. Augier, U. Boscain, and M. Sigalotti, “Adiabatic ensemble control of a continuum of quantum systems,” SIAM J. Control Optim., vol. 56, no. 6, pp. 4045–4068, 2018. [Online]. Available: https://doi.org/10.1137/17M1140327

[6] ——, “Semi-conical eigenvalue intersections and the ensemble controllability problem for quantum systems,” Math. Control Relat. Fields, vol. 10, no. 4, pp. 877–911, 2020. [Online]. Available: https://doi.org/10.3934/mcrf.2020023

[7] Z. Leghtas, A. Sarlette, and P. Rouchon, “Adiabatic passage and ensemble control of quantum systems,” Journal of Physics B, vol. 44, no. 15, 2011.

[8] F. C. Chittaro and J.-P. Gauthier, “Asymptotic ensemble stabilizability of the Bloch equation,” Systems Control Lett., vol. 113, pp. 36–44, 2018. [Online]. Available: https://doi.org/10.1016/j.sysconle.2018.01.008

[9] U. A. Maciel Neto, P. S. Pereira da Silva, and P. Rouchon, “Motion planing for an ensemble of Bloch equations towards the south pole with smooth bounded control,” Automatica J. IFAC, vol. 145, pp. Paper No. 110 529, 9, 2022. [Online]. Available: https://doi.org/10.1016/j.automatica.2022.110529

[10] R. Tycko, “Broadband population inversion,” Phys. Rev. Lett., vol. 51, pp. 775–777, Aug 1983. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevLett.51.775

[11] U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann, “Population transfer between molecular vibrational levels by stimulated Raman scattering with partially overlapping laser fields. a new concept and experimental results,” The Journal of Chemical Physics, vol. 92, no. 9, pp. 5363–5376, 1990.

[12] U. Boscain, G. Charlot, J.-P. Gauthier, S. Guérin, and H.-R. Jauslin, “Optimal control in laser-induced population transfer for two-and three-level quantum systems,” Journal of Mathematical Physics, vol. 43, no. 5, pp. 2107–2132, 2002.

[13] K. Bergmann, H.-C. Nägerl, C. Panda, G. Gabrielse, E. Miloglyadov, M. Quack, G. Seyfang, G. Wichmann, S. Ospelkaus, A. Kuhn, et al., “Roadmap on STIRAP applications,” Journal of Physics B: Atomic, Molecular and Optical Physics, vol. 52, no. 20, p. 202001, 2019.

[14] U. Boscain, M. Caponigro, T. Chambrion, and M. Sigalotti, “A weak spectral condition for the controllability of the bilinear Schrödinger equation with application to the control of a rotating planar molecule,” Comm. Math. Phys., vol. 311, no. 2, pp. 423–455, 2012. [Online]. Available: https://doi.org/10.1007/s00220-012-1441-z

[15] B. C. Hall, “An elementary introduction to groups and representations,” 2000.