

Trap-free manipulation in the Landau-Zener system

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The analysis of traps, that is, locally but not globally optimal controls, for quantum control systems has attracted great interest in recent years. The central problem that has remained open is to demonstrate for a given system either the existence or the absence of traps. We prove the absence of traps and hence completely solve this problem for the important tasks of unconstrained manipulation of the transition probability and unitary gate generation in the Landau-Zener system, a system with a wide range of applications across physics, chemistry, and biochemistry. This finding provides an example of a controlled quantum system which is completely free of traps. We also discuss the impact of laboratory constraints due to decoherence, noise in the control pulse, and restrictions on the available controls, which, when being sufficiently severe, can produce traps.

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I. INTRODUCTION

Manipulation by atomic and molecular systems is an important branch of modern science with applications ranging from optimal laser-driven population transfer in atomic systems to laser-assisted control of chemical reactions [1]. Much interest is directed towards control of the Landau-Zener (LZ) system, a two-state quantum system whose unitary evolution under the action of the control $\varepsilon(t)$ (e.g., shaped laser field) is governed by the equation

$$\dot{U}_t^\varepsilon = -i[\Delta\sigma_x + \varepsilon(t)\sigma_z]U_t^\varepsilon, \quad U_{t=0}^\varepsilon = \mathbb{I}, \quad (1)$$

where $\Delta > 0$, and σ_x and σ_z are the Pauli matrices. The case $\varepsilon(t) = \varepsilon t$ with constant ε was studied by Landau, Zener, Stückelberg, and Majorana [2]. This system has been widely applied in physics, chemistry, and biochemistry, for example, for describing transfer of charge along with its energy [3], photosynthesis [4], atomic and molecular collisions, processes in plasma physics [5], Bose-Einstein condensate [6], experimental realizations of qubits, etc. [7–13].

Controlled manipulation by a quantum system can be formulated as finding global maxima of a suitable objective $J(\varepsilon)$ associated to the system. For example, maximizing the probability of transition from the initial state $|i\rangle$ to a target final state $|f\rangle$ at a final time T can be described by maximizing $J(\varepsilon) = P_{i \rightarrow f} = |\langle f|U_T^\varepsilon|i\rangle|^2$. A control which attains a local maximum of J can be found either numerically using the model of the system or experimentally. In both circumstances, the first step of a common procedure is to apply a trial pulse ε_0 and obtain the outcome $J(\varepsilon_0)$, either numerically or measuring it in the laboratory. The second step is to make various small modifications of ε_0 and find ε_1 , which produces maximum increase in J . Then ε_1 is used as a new trial pulse and the procedure is repeated until no significant increase is produced or a maximum number of iterations is reached.

Of crucial practical importance is to know whether $J(\varepsilon)$ has *traps*, that is, local maxima with the values less than the global maximum, as necessary to properly choose between

local (e.g., gradient) and global optimization methods [14–19]. Traps can strongly influence both theoretical and experimental quantum control studies; they determine the level of difficulty of controlling the system and can significantly slow down or even completely prevent finding globally optimal controls. Whereas the analysis of traps in manipulation by quantum systems has attracted high attention [20–28], no examples of trap-free quantum systems have been known. Only partial theoretical results have been obtained, stating the absence of traps at special regular controls. This finding does not at all exclude the absence of traps that makes the problem open since even a single trap may produce significant difficulties for the optimization if it has a large attracting domain [29]. In this work we show that the LZ system is trap-free and hence, for example, the only extrema of $J(\varepsilon) = P_{i \rightarrow f}$ for this system are global maxima and minima. This finding provides an example of a trap-free quantum control system where unconstrained local manipulations are always sufficient to find best control pulses. Sufficiently strong constraints on the controls may destroy this property and in the end we discuss possible limitations for the analysis due to decoherence, noise in the control pulses, and limited tunability of the control strength and time scales in laboratory experiments.

II. TRAPS AND CONTROL LANDSCAPES

Formally, a control field $\varepsilon(t)$ is a trap for the objective $J(\varepsilon)$ if it is a local maximum, that is, a maximum with the value less than the global maximum, $J(\varepsilon) < J_{\max} = \max_{\varepsilon} J(\varepsilon)$ (in this work we consider as a control goal maximizing the objective; if the goal is to minimize the objective, then traps are local minima). Answering the question whether traps exist for a given control problem is crucial for determining proper algorithms and our abilities for finding optimal control fields. In the absence of traps, local search algorithms should generally be able to find globally optimal controls (exceptions may occur if the initial control is chosen exactly at a saddle point, where the gradient of the objective is zero). If the objective has traps (perhaps even a single trap with large attracting domain), then local search procedures may converge to local maxima instead of attaining a desired globally optimal

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control and more sophisticated global search methods should be exploited for a successful optimization.

Traps are critical points, that is, the gradient $\nabla J_\varepsilon = 0$ at any trap. Critical points for control objectives $J(\varepsilon) = P_{i \rightarrow f}$ were studied in seminal works [20], where the absence of traps was suggested. The suggestion was drawn from the proof that any function of the form $f(U) = \text{Tr}[U\rho_0 U^\dagger O]$ (ρ_0 is a positive matrix and O is Hermitian) defined on the unitary group $U(n)$, where n is the system dimension, has as extrema only global maxima, global minima, and saddles and has no traps. (Extrema of trace functions over unitary and orthogonal groups were studied in other contexts by von Neumann [30], Brockett [31], Glaser *et al.* [32], etc.) Then, under the controllability condition which assumes that any $U \in U(n)$ can be generated by some control, this result was used to conclude the absence of traps for the underlying objective functional $J(\varepsilon)$. Later it was shown that the conclusion of the absence of traps requires an additional assumption that the map $\chi : \varepsilon \rightarrow U_T^\varepsilon$ is nondegenerate [21], meaning that arbitrary infinitesimal variations of ε produce variations of U_T^ε in all directions on $U(n)$ [33,34]. While the controllability condition is relatively easy to verify [35], checking the nondegeneracy assumption turned out to be a hard problem. Moreover, critical controls violating this assumption were found [24,25], and even second-order traps—critical controls which are not global maxima and where the Hessian $H = \delta^2 J / (\delta\varepsilon)^2$ is negative semidefinite were shown to exist under rather general assumptions [26]. (Second-order traps are not necessarily local maxima but effectively they are traps for local algorithms exploiting at most second-order local information about the objective; see Chap. 20 of [36] for a general discussion of the nondegeneracy and second-order optimality conditions.) These findings led to reconsideration of the conclusion of an absence of traps. Some numerical simulations suggested that the condition of nondegeneracy might be generally satisfied or at least its violation does not produce multiple traps [27], while others indicated possible trapping behavior [25,29]. However, numerical search is limited and the extent to which these runs span the full space of quantum control possibilities is questionable [29]. Hence, the problem of proving either the existence or the absence of traps has remained open.

III. ABSENCE OF TRAPS FOR THE LANDAU-ZENER SYSTEM

Our main result is that the only critical points of any objective of the form $J(\varepsilon) = f(U_T^\varepsilon)$, where U_T^ε satisfies (1) and $f(U)$ is any function on the special unitary group $SU(2)$, which has no local extrema, are global maxima, global minima, and the zero control field $\varepsilon(t) = 0$. Important examples of such objectives include the following.

(i) Transition probability:

$$J_{i \rightarrow f}(\varepsilon) = |\langle f | U_T^\varepsilon | i \rangle|^2.$$

This objective is maximized by a control which completely transfers the initial state $|i\rangle$ into the desired final state $|f\rangle$.

(ii) Expectation of a system observable O :

$$J_O(\varepsilon) = \text{Tr}[U_T^\varepsilon \rho_0 U_T^{\varepsilon\dagger} O].$$

Here O is a Hermitian matrix representing the observable and ρ_0 is the initial system density matrix. The objective is maximized by a control which maximizes quantum-mechanical average of O at time T .

(iii) Generation of a unitary process W :

$$J_W(\varepsilon) = \frac{1}{4} |\text{Tr}(W^\dagger U_T^\varepsilon)|^2.$$

Here W is the unitary matrix representing a desired system evolution or a desired quantum gate, for example, Hadamard gate. The maximum of this objective is achieved by a control such that $U_T = e^{i\phi} W$, where ϕ is an arbitrary (generally unphysical) phase. Factor 1/4 is chosen to have $\max_\varepsilon J_W(\varepsilon) = 1$.

Proof of the main result. We consider first $J(\varepsilon) = J_{i \rightarrow f}(\varepsilon)$. For brevity, we sometimes omit the superscript ε in U_t^ε and U_T^ε , and without loss of generality set $\Delta = 1$. Gradient of $J(\varepsilon) = |\langle f | U_T^\varepsilon | i \rangle|^2$ for the LZ system has the form [26]

$$\nabla J_\varepsilon(t) = 2\text{Im}(\langle i | U_T^\dagger | f \rangle \langle f | U_T U_t^\dagger \sigma_z U_t | i \rangle). \quad (2)$$

It can be written as $\nabla J_\varepsilon(t) = L(U_t^\dagger \sigma_z U_t) = l(t)$, where $L : \text{su}(2) \rightarrow \mathbb{R}$ is the linear map on the Lie algebra of traceless Hermitian 2×2 matrices defined by $L(A) = 2\text{Im}[\langle i | U_T^\dagger | f \rangle \langle f | U_T A | i \rangle]$ and $l(t)$ is a real-valued function. If ε is a critical control field, then $l(t) \equiv 0$ and therefore, in particular, $l'(t) = l''(t) = 0$. These derivatives can be computed to be

$$\begin{aligned} l'(t) &= L(-iU_t^\dagger[\sigma_x + \varepsilon(t)\sigma_z, \sigma_z]U_t) = -2L(U_t^\dagger \sigma_y U_t), \\ l''(t) &= -2L(-iU_t^\dagger[\sigma_x + \varepsilon(t)\sigma_z, \sigma_y]U_t) \\ &= -4L(U_t^\dagger \sigma_z U_t) + 4\varepsilon(t)L(U_t^\dagger \sigma_x U_t). \end{aligned}$$

Thus, the condition $l'' = l' = l = 0$ for any t such that $\varepsilon(t) \neq 0$ takes the form

$$L(U_t^\dagger \sigma_x U_t) = L(U_t^\dagger \sigma_y U_t) = L(U_t^\dagger \sigma_z U_t) = 0. \quad (3)$$

The matrices $U_t^\dagger \sigma_x U_t$, $U_t^\dagger \sigma_y U_t$, $U_t^\dagger \sigma_z U_t$ are linearly independent traceless Hermitian 2×2 matrices. They form a basis of $\text{su}(2)$ and hence (3) implies $L(A) = 0$ for any $A \in \text{su}(2)$.

Let $|i_\perp\rangle$ be the state which is orthogonal to $|i\rangle$. Taking $A = |i\rangle\langle i_\perp| + |i_\perp\rangle\langle i|$ and $A' = i(|i\rangle\langle i_\perp| - |i_\perp\rangle\langle i|)$ gives

$$\begin{aligned} L(A) &= 0 \Rightarrow \text{Im}(\langle i | U_T^\dagger | f \rangle \langle f | U_T | i_\perp \rangle) = 0, \\ L(A') &= 0 \Rightarrow \text{Re}(\langle i | U_T^\dagger | f \rangle \langle f | U_T | i_\perp \rangle) = 0. \end{aligned}$$

Thus, $\langle i | U_T^\dagger | f \rangle \langle f | U_T | i_\perp \rangle = 0$; that is, either $\langle i | U_T^\dagger | f \rangle = 0$ or $\langle f | U_T | i_\perp \rangle = 0$. The former case corresponds to the global minimum of the objective ($J = 0$) and the latter to its global maximum ($J = 1$). These are the only allowed critical controls except of $\varepsilon(t) \equiv 0$. This finishes the proof of the main result for $J_{i \rightarrow f}(\varepsilon)$.

The analysis above immediately implies that if a linear map $L : \text{su}(2) \rightarrow \mathbb{R}$ satisfies $L(U_t^\dagger \sigma_z U_t) = 0$ then $L \equiv 0$. Now we show that it means that the map $\chi : \varepsilon \rightarrow U_T^\varepsilon$ is nondegenerate everywhere outside of $\varepsilon(t) \equiv 0$. Since we consider objectives produced by functions on $SU(2)$ which are therefore invariant with respect to the overall phase of U_T^ε , we can identify U_T^ε with the corresponding element of $SU(2)$. Small variations around U_T^ε can be represented as $\tilde{U}_T^\varepsilon = U_T^\varepsilon e^{\delta w} \approx U_T^\varepsilon (1 + \delta w)$,

where $\delta w = -i \int_0^T U_t^\dagger \sigma_z U_t \delta \varepsilon(t) dt$. For the map χ to be non-degenerate, \tilde{U}_T^ε should span a neighborhood of U_T^ε that in turn requires δw to span $\mathfrak{su}(2)$. If δw does not span $\mathfrak{su}(2)$, then there exists $A \in \mathfrak{su}(2)$, $A \neq 0$ such that $(A, \delta w) \equiv \text{Tr}(A^\dagger \delta w) = 0$ for all $\delta \varepsilon$ and hence $L_A(U_t^\dagger \sigma_z U_t) := \text{Tr}(A^\dagger U_t^\dagger \sigma_z U_t) = 0$. This is possible only if $A = 0$ and hence the map cannot be degenerate. Therefore, our result immediately implies the absence of traps at any $\varepsilon \neq 0$ for any objective functional $J(U_T^\varepsilon)$ which has no traps if considered as a function on $\text{SU}(2)$. This includes important objectives $J_O = \text{Tr}[U_T^\varepsilon \rho_0 U_T^{\varepsilon\dagger} O]$ for maximizing expectation of a system observable O and $J_W = (1/4)|\text{Tr}(W^\dagger U_T^\varepsilon)|^2$ for optimal generation of a unitary process W (e.g., for unitary gate generation). These objectives appear to be trap-free for the LZ system since functions $f_O(U) = \text{Tr}[U \rho_0 U^\dagger O]$ and $f_W(U) = (1/4)|\text{Tr}(W^\dagger U)|^2$ have no local maxima on $\text{SU}(2)$ [20].

The control $\varepsilon(t) \equiv 0$ requires a separate consideration since the condition $l''(t) = 0$ for $\varepsilon \equiv 0$ cannot be used to conclude $L(U_t^\dagger \sigma_x U_t) = 0$. This control is, however, not a trap, for example, for $J_{i \rightarrow f}$ as shown by direct computation in the Appendix.

IV. DISCUSSION

Now we discuss important limitations for the present analysis. No real-world system will perfectly evolve according to Eq. (1) and three general kinds of deviations from the ideal situation include decoherence effects, deviations of the actual control from the intended one due to noise or imperfections of the laboratory setup, and limited tunability of the control strength and time scales in laboratory experiments. While we consider the system as evolving according to the Schrödinger equation with unitary evolution, in real circumstances it can experience additional influence of the environment which causes the dynamics to be nonunitary. We also assume that any shape of the control $\varepsilon(t)$ is available, whereas typical pulses are either piecewise constant or finite sums of cosines and sines at certain fixed frequencies. These assumptions are common for the first step of control landscape analysis which deals with the ideal situation of noiseless unconstrained controls. The next step upon establishment of the ideal landscape properties is to study the effects of possible deviations, which we discuss below for the LZ system.

The requirements on the available control fields (e.g., on their strength and time scale) necessary for the conclusion of the absence of traps for attaining maximal objective value are such that the available controls are sufficient to guarantee controllability of the system. Minimal control time for the LZ system can be estimated using the fundamental theory of optimal control at the quantum speed limit as $T_{\text{QSL}} \approx \Delta E_0^{-1} \arccos(|\langle i, f \rangle|)$, where ΔE_0 is the energy variance of the free Hamiltonian $H_0 = \Delta \sigma_x$ calculated on the initial state [10]. Hence, our analysis applies to any final time $T \gtrsim \pi \Delta E_0^{-1}$. If for a given physical system decoherence effects occur on a time scale slower than ΔE_0^{-1} , they can be neglected when the control is implemented in the time optimal fashion. This shows that while finite-time [37] and decoherence [38–40] effects can be important for the LZ system, they do not modify the trap-free landscape property as soon as final time T is

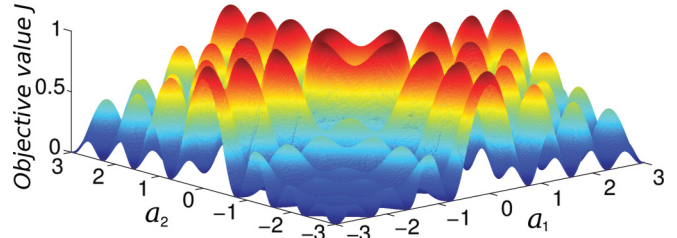


FIG. 1. (Color online) The control landscape of $J_{0 \rightarrow 1}(a_1, a_2)$ for the LZ system controlled by piecewise constant controls ($N = 2$, $T = 10$, $\Delta = 1$). The landscape possesses multiple traps (local maxima).

sufficiently smaller than the relaxation time and at the same time is not too small to violate controllability of the systems.

An extensive numerical analysis of control landscapes for multilevel model systems with realistic laboratory control fields is provided in [27]. To analyze the role of limitations on the available control fields for the LZ system, we numerically estimate the probability of trapping when available controls are piecewise constant controls of the form $\varepsilon(t) = \sum_{i=1}^N a_i \chi_{[t_i, t_{i+1})}(t)$, where $\chi_{[t_i, t_{i+1})}(t) = 1$ if $t \in [t_i, t_{i+1})$ and zero otherwise and a_i are the control parameters. Typically, $N \approx 100$ and control amplitudes are constrained within certain ranges, say, $a_i \in [-A, A]$. An exact solution for piecewise constant controls can be obtained, for example, in the simplest case $N = 1$. The objective for maximizing the probability of spin flip $J_{0 \rightarrow 1} = |\langle 0 | U_T^\varepsilon | 1 \rangle|^2$ by a constant control $\varepsilon(t) = a$ can be computed to be $J_{0 \rightarrow 1}(a) = \sin^2(T\sqrt{1+a^2})/(1+a^2)$. Its traps (local maxima) are given by solutions of the equation $\tan(T\sqrt{1+a^2}) = T\sqrt{1+a^2}$; the corresponding objective values are $J_{0 \rightarrow 1}(a) = T^2/(1+T^2+T^2a)$. Control landscapes for a two-dimensional control space ($N = 2$) are more complex. Figure 1 shows as an example the control landscape of $J_{0 \rightarrow 1}(a_1, a_2)$. The landscape has multiple local maxima showing that significant restrictions on the control space in an originally trap-free system may produce traps. Figure 2 provides the numerically estimated probability of trapping for piecewise constant controls as a function of N . The probability of trapping becomes negligible already for $N = 10$ –15, which means that limitations on the number of components N of available laboratory control fields have minor effect already for $N \gtrsim 10$ and hence should be negligible for the realistic case $N \approx 100$.

In the laboratory, actual controls may deviate from the designed numerically optimal pulse due to noise and imperfections of experimental setup [42]. These noise effects can influence on the landscape structure by decreasing the maximal objective value. We adopt the general theory of [43] to analyze this influence for the LZ system. Let $\varepsilon_0(t)$ be an optimal control in the ideal situation of absence of noise. In the presence of a random noise $\xi(t)$, the actual control will fluctuate as $\varepsilon(t) = \varepsilon_0(t) + \varrho(t)\xi(t)$, where $\varrho(t) = 1$ for additive noise and $\varrho(t) = \varepsilon_0(t)$ for multiplicative noise. A weak noise modifies the averaged objective as

$$\begin{aligned} \mathbb{E}[J(\varepsilon_0)] \\ \approx J(\varepsilon_0) + \frac{1}{2} \int_0^T \int_0^T \mathcal{H}_0(t, t') \varrho(t) \varrho(t') \mathbb{E}[\xi(t) \xi(t')] dt dt', \end{aligned}$$

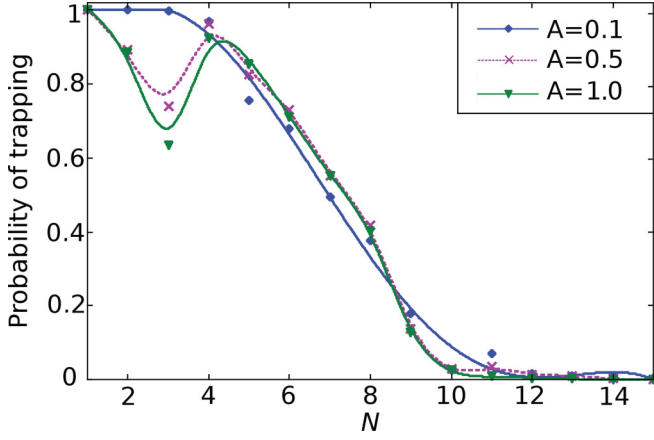


FIG. 2. (Color online) Probability of trapping as a function of N for piecewise constant controls ($T = 10$, $\Delta = 1$). For every point, 10^3 runs of MATLAB realization of the Broyden-Fletcher-Goldfarb-Shanno (BFGS) optimization algorithm were performed each starting at a random initial control $a = (a_1, \dots, a_N)$ [41]. Initial control amplitudes are uniformly distributed in the range $a_i \in [A, A]$ but are allowed to escape this range during the search. The search is defined as trapped if the attained objective is less than 0.99. Trapping may occur due to the presence of local maxima and/or principal impossibility of attaining the objective value greater than 0.99 with available controls. Probability of trapping is estimated as a fraction of trapped runs among all 10^3 runs.

where $\mathcal{H}_0(t, t') = \frac{\delta^2 J}{\delta \varepsilon_0(t) \delta \varepsilon_0(t')}$ is the Hessian of the objective computed at the optimal control field ε_0 and $\mathbb{E}[\xi(t)\xi(t')]$ is the autocorrelation function of the noise. Since Hessian is negative semidefinite at the maximum, the noise generally decreases the average fidelity. The objective for additive (AWN) and multiplicative (MWN) white noise with autocorrelation function $\mathbb{E}[\xi(t)\xi(t')] = \sigma \delta(t - t')$, where σ^2 is the variance of the noise amplitude distribution, take the forms

$$\mathbb{E}_{\text{AWN}}[J(\varepsilon_0)] \approx J(\varepsilon_0) + \frac{\sigma^2}{2} \int_0^T \mathcal{H}_0(t, t) dt,$$

$$\mathbb{E}_{\text{MWN}}[J(\varepsilon_0)] \approx J(\varepsilon_0) + \frac{\sigma^2}{2} \int_0^T \mathcal{H}_0(t, t) |\varepsilon_0(t)|^2 dt.$$

The last term in these equations is the noise-induced decrease $-\mathcal{D}(\varepsilon_0, \sigma, T)$ of the objective [such that $\mathbb{E}(J) \approx J(\varepsilon_0) - \mathcal{D}$ with $\mathcal{D} \geq 0$]. The diagonal of the Hessian for $J = J_{i \rightarrow f}$ can be shown to be $\mathcal{H}_0(t, t) = -2|\langle i | U_t^\dagger \sigma_z U_t | i_\perp \rangle|^2$ so that $|\mathcal{H}_0(t, t)| \leq 2$. Therefore, $\mathcal{D}(\varepsilon_0, \sigma, T)$ for $J = J_{i \rightarrow f}$ is majorized by

$$\mathcal{D}_{\text{AWN}}(\varepsilon_0, \sigma, T) \leq \sigma^2 T, \quad \mathcal{D}_{\text{MWN}}(\varepsilon_0, \sigma, T) \leq \sigma^2 E,$$

where $E = \int_0^T |\varepsilon_0(t)|^2 dt$ is the total energy of the pulse. The diagonal of the Hessian for the objective J_W is $\mathcal{H}_0(t, t) = -2$ and therefore for this objective $\mathcal{D}_{\text{AWN}} = \sigma^2 T$ and $\mathcal{D}_{\text{MWN}} = \sigma^2 E$. It then follows that in both cases the influence of a weak AWN can be minimized by using time optimal controls, while minimizing weak MWN can be done by selecting less energetic pulses among all optimal pulses.

If the ideal landscape has multiple global optima with different $\mathcal{H}_0(t, t)$, then the noise-induced decrease of objective can be different at different optima that can produce traps in the nonideal landscape. Weak decoherence operates similarly to

weak noise and can also produce traps in the ideally trap-free landscape [20]. These deviations from the ideal situation should be avoided to reveal the trap-free landscape property by either operating in the time optimal regime or using weak optimal controls to combat MWN. Strong noise and strong decoherence that can significantly modify the landscape are beyond the scope of this discussion.

V. CONCLUSIONS

This work shows that unconstrained manipulation in the LZ system is free of traps and hence unconstrained local search for optimal controls is always able to find the best optima. The impact on this result of laboratory limitations due to decoherence, noise in the actual control pulses, and restrictions on the available control fields is discussed.

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APPENDIX

Here we prove that the control $\varepsilon(t) \equiv 0$ is not a trap for state-to-state transfer described by the objective $J(\varepsilon) = J_{i \rightarrow f}(\varepsilon)$. The evolution operator produced by $\varepsilon(t) = 0$ has the form $U_t = e^{-it\sigma_x}$. Therefore, $V_t := U_t^\dagger \sigma_z U_t = \cos(2t)\sigma_z + \sin(2t)\sigma_y$ and the gradient of the objective is

$$\nabla J_{\varepsilon=0}(t) = \cos 2t L(\sigma_z) + \sin 2t L(\sigma_y).$$

If $\varepsilon(t) = 0$ is a critical point, then $\nabla J_{\varepsilon=0}(t) = 0$ for any $t \in [0, T]$, and hence $L(\sigma_z) = L(\sigma_y) = 0$. If $\alpha := L(\sigma_x) = 0$, then $L \equiv 0$ on $\mathfrak{su}(2)$ and similarly to the proof of the main result we conclude that $\varepsilon = 0$ is not a trap.

Now consider the case $\alpha \neq 0$. In this case $|i\rangle$ and $|f\rangle$ are such that $\varepsilon = 0$ is neither a global maximum nor global minimum. The evolution operator produced by a small variation of the control $\delta\varepsilon$ can be represented as $U_T^{\delta\varepsilon} = e^{-iT\sigma_x} W_T$, where W_T satisfies

$$\dot{W}_t = -i\delta\varepsilon(t)V_t W_t, \quad W_0 = \mathbb{I}.$$

The operator W_T can be computed up to the second order in $\delta\varepsilon$ as

$$W_T = \mathbb{I} + A_1 + A_2 + o(\|\delta\varepsilon\|^2), \quad A_1 = -i \int_0^T dt \delta\varepsilon(t) V_t,$$

$$A_2 = - \int_0^T dt_1 \int_0^{t_1} dt_2 \delta\varepsilon(t_1) \delta\varepsilon(t_2) V_{t_1} V_{t_2}.$$

This gives the perturbation expansion for the objective (here $|f'\rangle = e^{iT\sigma_z}|f\rangle$),

$$J(\delta\varepsilon) = |\langle f' | \mathbb{I} + A_1 + A_2 + \dots | i \rangle|^2$$

$$= |\langle f' | i \rangle|^2 + \delta J_1(\delta\varepsilon) + \delta J_2(\delta\varepsilon) + o(\|\delta\varepsilon\|^2),$$

where

$$\begin{aligned}\delta J_1(\delta\varepsilon) &= 2\text{Re}(\overline{\langle f'|i\rangle}\langle f'|A_1|i\rangle), \\ \delta J_2(\delta\varepsilon) &= |\langle f'|A_1|i\rangle|^2 + 2\text{Re}(\overline{\langle f'|i\rangle}\langle f'|A_2|i\rangle).\end{aligned}$$

Hence, the variation of the objective satisfies [note that $|\langle f'|i\rangle|^2 = J(0)$]

$$\delta J = J(\delta\varepsilon) - J(0) = \delta J_1(\delta\varepsilon) + \delta J_2(\delta\varepsilon) + o(\|\delta\varepsilon\|^2).$$

If $\varepsilon = 0$ is a critical control, then $\delta J_1(\delta\varepsilon) = 0$ for any $\delta\varepsilon$. We will show the existence of controls $\delta\varepsilon_1$ and $\delta\varepsilon_2$ such that $\delta J_2(\delta\varepsilon_1)$ and $\delta J_2(\delta\varepsilon_2)$ have opposite signs. It is sufficient to choose $\delta\varepsilon_1$ and $\delta\varepsilon_2$ to satisfy $\langle f'|A_1|i\rangle = 0$, for example,

$$\int_0^T dt \delta\varepsilon_i(t) \cos 2t = \int_0^T dt \delta\varepsilon_i(t) \sin 2t = 0, \quad i = 1, 2.$$

Since $V_{t_1} V_{t_2} = \cos 2(t_1 - t_2) + i\sigma_x \sin 2(t_1 - t_2)$, we have

$$\begin{aligned}\delta J_2(\delta\varepsilon) &= 2 \int_0^T dt_1 \int_0^{t_1} dt_2 \delta\varepsilon(t_1) \delta\varepsilon(t_2) (J(0) \cos 2(t_1 - t_2) \\ &\quad + \alpha \sin 2(t_1 - t_2)) \\ &= 2\alpha \int_0^T dt_1 \int_0^{t_1} dt_2 \delta\varepsilon(t_1) \delta\varepsilon(t_2) \sin 2(t_1 - t_2).\end{aligned}$$

Assuming for simplicity that $T \geq \pi$, we take $\delta\varepsilon_1(t) = \chi_{[0,\pi]}(t)$ and $\delta\varepsilon_2(t) = \cos(4t)\chi_{[0,\pi]}(t)$, where $\chi_{[0,\pi]}(t)$ is the characteristic function of the interval $[0,\pi]$. Then $\delta J_2(\delta\varepsilon_1) = -\pi\alpha$ and $\delta J_2(\delta\varepsilon_2) = \pi\alpha/6$. Therefore for $\alpha \neq 0$ there exist control variations around $\varepsilon(t) = 0$ increasing the objective and control variations decreasing it. This implies that $\varepsilon(t) = 0$ is neither a local maximum nor minimum.

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