Quantum control landscape for a Λ-atom in the vicinity of second order traps

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Abstract

We show that the second order traps in the control landscape for a three-level Λ-system found in our previous work Phys. Rev. Lett. 106, 120402 (2011) are not local maxima: there exist directions in the space of controls in which the objective grows. The growth of the objective is slow — at best 4th order for weak variations of the control. This implies that simple gradient methods would be problematic in the vicinity of second order traps, where more sophisticated algorithms that exploit the higher order derivative information are necessary to climb up the control landscape efficiently. The theory is supported by a numerical investigation of the landscape in the vicinity of the ε(t) = 0 second order trap, performed using the GRAPE and BFGS algorithms.

1 Introduction

Manipulation of atomic systems via specially tailored laser pulses is an important application of quantum control theory [1, 2, 3, 4, 5, 6, 7, 8, 9]. One of the questions of current interest is the nature of the control landscape that describes the value of the objective as a functional of the control field ε(t), in particular the nature of the critical points on the landscape. For the objective J(ε) a critical control field ε is a trap of the landscape if ε is a local but not a global maximum. Control landscapes for closed quantum systems were studied in [10, 11], where the absence of traps was shown under some assumptions. The analysis for open quantum systems was performed in [12, 13] and later was extended to a unified analysis of control landscapes for open classical and quantum systems [14].

In this work we consider critical points for control objectives of the form

\[ J(\varepsilon) = \text{Tr}[U_T^\varepsilon \rho_0 U_T^{\dagger \varepsilon} O] \] (1)

for a 3-level Λ-system isolated from the environment. Such objectives describe a wide variety of quantum control phenomena, e.g., breaking a desired chemical bond, producing selective...
atomic and molecular excitations, etc. Here \( \rho_0 = |i⟩⟨i| \) is the initial system state which is assumed to be pure, \( O \) is the target observable, and \( U_T^\varepsilon \) is the evolution operator describing the evolution of the system from the initial time \( t = 0 \) to the final time \( T > 0 \) under the action of coherent control field \( \varepsilon(t) \). The evolution operator satisfies the equation

\[
\frac{dU_T^\varepsilon}{dt} = -i(H_0 + V\varepsilon(t))U_T^\varepsilon, \quad U_0^\varepsilon = I
\]

where \( H_0 = \sum_{j=1}^{3} \varepsilon_j |j⟩⟨j| \) is the free system Hamiltonian and \( V \) is the operator describing coupling of the system to the control field.

A control field \( \varepsilon \) is a second order trap for the objective functional \( J(\varepsilon) \) if the gradient \( \nabla J(\varepsilon) = 0 \) and the Hessian \( \delta^2 J/\delta\varepsilon^2 \) is negative semi-definite but \( J(\varepsilon) \) is not a global maximum. More generally, a control field \( \varepsilon \) is an \( n \)-th order trap (here \( n \) is an even natural number) if it is not a global maximum and \( \delta J(\varepsilon) := J(\varepsilon + \delta\varepsilon) - J(\varepsilon) = R(\delta\varepsilon) + O(\delta\varepsilon^{n+1}) \), where the functional \( R \neq 0 \), \( R(\delta\varepsilon) = O(\delta\varepsilon^n) \), and \( R(\delta\varepsilon) \leq 0 \).

Various algorithms have been used to find optimal controls in quantum systems including gradient search, e.g. GRAPE [15] and its modifications for rapidly time-varying Hamiltonians [16], Krotov-type methods [17, 18, 19, 20, 21], the Broyden-Fletcher-Goldfarb-Shanno (BGFS) algorithm and its modifications [22, 23, 24, 25], genetic algorithms and evolutionary strategies [26, 27], and combined approaches [28, 29]. The analysis of the existence or absence of traps, including \( n \)-order traps, is important for determining proper algorithms for finding optimal control fields. In the absence of traps, local (e.g., gradient) search should generally be able to find globally optimal controls (exceptions may occur if the initial control for a gradient-based search is chosen exactly at a saddle point, where the gradient of the objective is zero). If the landscape has second order or \( n \)-order traps then, since the gradient of the objective at a critical point is zero and in addition, at second-order or \( n \)-order traps the Hessian is negative semi-definite, the objective in their vicinity may grow not faster than at a third order in the small variation \( \delta\varepsilon \) of the control. Therefore the search for globally optimal controls with simple gradient methods would be problematic in a vicinity of second-order or \( n \)-order traps, where more sophisticated algorithms exploiting the higher order information about the objective are necessary to efficiently climb up the control landscape.

Second order traps were shown to exist for a general class of quantum systems [30]. The simplest example is a three-level \( \Lambda \)-system, where zero control field was shown to be a second order trap. In this work we continue the analysis of [30] and show that this second order trap is not a local maximum; there exist a direction in which the objective grows. We also perform a numerical study of the landscape in a vicinity of the \( \varepsilon(t) = 0 \) second order trap.

## 2 Three-level \( \Lambda \)-system

The simplest example where second order traps appear is the following problem: maximizing the expectation of an operator \( O = \sum_{j=1}^{3} \lambda_j |j⟩⟨j| \) with \( \lambda_2 > \lambda_1 > \lambda_3 \) for a three-level non-degenerate \( \Lambda \)-atom which is initially in the ground state \( \rho_0 = |1⟩⟨1| \) (Fig. 1). The interaction Hamiltonian \( V \) for \( \Lambda \)-atom satisfies \( V_{12} = 0 \), which is consistent with the controllability assumption if \( V_{13} \neq 0 \) and \( V_{23} \neq 0 \). Note that the pure state controllability for \( N \)-level systems with odd number of levels requires producing the full unitary group \( su(N) \), while for
Figure 1: The simplest example of a quantum system possessing a second order trap is a three-level Λ-system initially in the ground state. The control field $\varepsilon(t) = 0$ is a second order trap for maximizing expectation of any target operator of the form $O = \sum_{i=1}^{3} \lambda_i |i\rangle \langle i|$ with $\lambda_2 > \lambda_1 > \lambda_3$. However, this field is not a trap, as shown in Theorem 2.

$N$ even the symplectic group $sp(N/2)$ alone is sufficient [31, 32]. The two allowed transition frequencies are $\omega_1 = \varepsilon_3 - \varepsilon_1$ and $\omega_2 = \varepsilon_3 - \varepsilon_2$; non-degeneracy implies that $\omega_1 \neq \omega_2$.

Globally optimal control fields are those which steer $|1\rangle$ completely into $|2\rangle$ producing the global maximum of the objective with the objective value $J_{\text{max}} = \lambda_2$.

Without loss of generality we can set $\lambda_1 = 0$ and $\lambda_2 = 1$ (replacing $O$ by $O' = (O - \lambda_1 I) / (\lambda_2 - \lambda_1)$ and noticing that linear transformations of the target observable do not affect the properties of critical points). Since $\lambda_3 < \lambda_1 = 0$, we can set $\lambda_3 = -\lambda$, where $\lambda > 0$.

After these transformations the objective $J$ takes the form

$$J(\varepsilon) = P_{1\rightarrow 2}(\varepsilon) - \lambda P_{1\rightarrow 3}(\varepsilon), \quad \lambda > 0$$

(2)

where $P_{i\rightarrow f}(\varepsilon) = |\langle f | U_T^\dagger |i\rangle|^2$ is the transition probability from the state $|i\rangle$ to the state $|f\rangle$ ($i = 1, f = 2, 3$). The objective takes the values in the interval $-\lambda \leq J \leq 1$.

If $\varepsilon(t) = 0$, then the system remains in the ground state and therefore both transition probabilities are zero, $P_{1\rightarrow 2}(0) = P_{1\rightarrow 3}(0) = 0$. The corresponding objective value $J = 0$ is neither a global maximum nor a global minimum.

**Theorem 1** The control field $\varepsilon(t) = 0$ is a second order trap with the objective value $J = 0 < J_{\text{max}}$.

More generally, a control $\varepsilon(t) = \varepsilon_0$ is a second order trap if the initial density matrix and target operator have the form $\rho_0 = |\tilde{k}\rangle \langle \tilde{k}|$ and $O = \sum_{i=1}^{n} \lambda_i |i\rangle \langle i|$ in the basis $|i\rangle$ of the modified Hamiltonian $\tilde{H}_0 = H_0 - \mu \varepsilon_0$, where $1 < k < n$ and $\lambda_1 > \lambda_2 > \ldots > \lambda_n$, and the dipole moment satisfies $\langle \tilde{i}| \mu |\tilde{k}\rangle = 0$ for all $i < k$. This important result was proved in [30]. However, previously it was not known if these second order traps are true traps (local maxima). The following theorem states that it is not the case and there exist local directions in which the objective increases. The analysis of the general case is equivalent to the case $\varepsilon(t) = 0$ with the modified Hamiltonian $\tilde{H}_0$, therefore only the case $\varepsilon(t) = 0$ will be treated in the rest of the manuscript.

**Theorem 2** The control field $\varepsilon(t) = 0$ is not a local maximum; there exists a direction $\delta \varepsilon$ in which the objective $J(\delta \varepsilon)$ grows as $J \approx \|\delta \varepsilon\|^4$. 

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**Proof.** Matrix elements determining the transition probabilities under the action of a small $\delta \varepsilon$ can be expanded in powers of $\delta \varepsilon$ as

\[
\langle 3 | U^\delta \varepsilon_T | 1 \rangle = A_1 + A_2 + A_3 + O(\delta \varepsilon^4) \tag{3}
\]
\[
\langle 2 | U^\delta \varepsilon_T | 1 \rangle = B_2 + O(\delta \varepsilon^3) \tag{4}
\]

where the terms $A_n$ and $B_n$ are of the order $O(\delta \varepsilon^n)$. Their explicit form can be derived from the perturbation expansion

\[
U^\delta \varepsilon_T = e^{-iTH_0} \left( \mathbb{I} + \sum_{n=1}^{\infty} (-i)^n \int_0^T dt_1 \int_0^{t_1} dt_2 \ldots \int_0^{t_{n-1}} dt_n \delta \varepsilon(t_1) \ldots \delta \varepsilon(t_n)V_{t_1} \ldots V_{t_n} \right)
\]

for the evolution operator $U^\delta \varepsilon_T$ (where $V_t = e^{itH_0}V e^{-itH_0}$) as:

\[
A_1 = -ie^{-iT\varepsilon_3} \int_0^T dt_1 \delta \varepsilon(t_1) \langle 3 | V_{t_1} | 1 \rangle = -iV_{31} e^{-iT\varepsilon_3} \int_0^T dt_1 \delta \varepsilon(t_1) e^{it\omega_1} \tag{5}
\]
\[
A_2 = -e^{-iT\varepsilon_3} \int_0^T dt_1 \int_0^{t_1} dt_2 \delta \varepsilon(t_1) \delta \varepsilon(t_2) \langle 3 | V_{t_1} V_{t_2} | 1 \rangle \tag{6}
\]
\[
A_3 = ie^{-iT\varepsilon_3} \int_0^T dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \delta \varepsilon(t_1) \delta \varepsilon(t_2) \delta \varepsilon(t_3) \langle 3 | V_{t_1} V_{t_2} V_{t_3} | 1 \rangle \tag{7}
\]
\[
B_2 = -e^{-iT\varepsilon_2} \int_0^T dt_1 \int_0^{t_1} dt_2 \delta \varepsilon(t_1) \delta \varepsilon(t_2) \langle 2 | V_{t_1} V_{t_2} | 1 \rangle \tag{8}
\]

Note that $\langle 3 | V_{t_1} V_{t_2} | 1 \rangle = 0$ and thus $A_2 = 0$ for any $\delta \varepsilon$. Substituting eqs. (3) and (4) into eq. (2) produces the following expansion for the objective around zero control field:

\[
\delta J_{\varepsilon=0} = J(\delta \varepsilon) - J(0) = -\lambda |A_1|^2 + \left( |B_2|^2 - 2\lambda \text{Re}(A_1^* A_3) \right) + O(\delta \varepsilon^6)
\]

(If the diagonal elements of $V$ are zero as is normally the case for the $\Lambda$-system, this implies that for the $\Lambda$-system $\langle 2 | V_{t_1} V_{t_2} V_{t_3} | 1 \rangle = 0$ and $\langle 3 | V_{t_1} V_{t_2} V_{t_3} V_{t_4} | 1 \rangle = 0$. Therefore the term of fifth order on the right hand side of this equation vanishes and the remainder is of sixth order.)

If $A_1 \neq 0$, then $\delta J = -\lambda |A_1|^2 + O(\delta \varepsilon^4) < 0$ for sufficiently small $\delta \varepsilon$. The Hessian of the objective at $\varepsilon = 0$ is negative semi-definite and therefore $\varepsilon = 0$ is a second order trap. However, if there would exist a $\delta \varepsilon$ such that $A_1 = 0$ and $B_2 \neq 0$, then would be $\delta J = |B_2|^2 + O(\delta \varepsilon^6) > 0$ and $\varepsilon = 0$ would not be a trap.

An example of $\delta \varepsilon$ for which $A_1 = 0$ and $B_2 \neq 0$ is the following (we assume $T \geq T' := 2\pi/\omega_1$). Set $\delta \varepsilon(t) = \alpha \chi_{[0,T']}(t)$, where $\chi_{[0,T]}$ is the characteristic function of the segment $[0,T]$, i.e. $\delta \varepsilon(t) = \alpha$ if $0 \leq t \leq T'$ and $\delta \varepsilon(t) = 0$ if $t > T'$ ($\alpha > 0$ is a small number). Then

\[
A_1 = \frac{V_{31}}{\omega_1} \left( 1 - e^{iT\omega_1} \right) e^{-iT\varepsilon_3} = 0
\]
\[
B_2 = -V_{23} V_{31} \int_0^{T'} dt_1 e^{it_1 \omega_1} \delta \varepsilon(t_1) \int_0^{t_1} dt_2 e^{-it_2 \omega_2} \delta \varepsilon(t_2)
\]
\[
= \alpha^2 V_{23} V_{31} e^{2\pi i r} - \frac{1}{\omega_2 (\omega_2 - \omega_1)}, \quad r = \frac{\omega_2}{\omega_1}
\]
The quantity $B_2 \neq 0$ if $r \notin \mathbb{Z}$ which is trivially satisfied since $0 < \omega_2 < \omega_1$. This proves the Theorem.

The most general conditions for $\delta e$ to increase the objective at the 4th order are

$$
\tilde{\delta}e_1(\omega_1) := \int_0^T dt e^{it\omega_1} \delta e(t) = 0
$$

$$
\tilde{\delta}e_2(\omega_1,\omega_2) := \int_0^T dt_1 e^{it_1\omega_1} \delta e(t_1) \int_0^{t_1} dt_2 e^{-it_2\omega_2} \delta e(t_2) \neq 0
$$

The condition $\tilde{\delta}e(\omega_1) = 0$ means that a control field $\delta e$ which increases the objective should have zero amplitude at the frequency $\omega_1$ of the $1 \to 3$ transition. Such a field does not populate the level $|3\rangle$ in the first order of the perturbation theory (via one-photon processes). Because of the special form of the interaction Hamiltonian $V$, the level $|3\rangle$ can not be populated also in the second order of the perturbation theory (via two-photon processes). However, level $|2\rangle$ can be populated at second order in the perturbation even by fields which have zero amplitude at the frequency $\omega_1$ if $\tilde{\delta}e_2(\omega_1,\omega_2) \neq 0$. Such a transfer of population increases the objective at the rate $\sim \delta e^4$. Similar control pulses which are non-resonant but have non-zero two-photon transition probability were used for coherent control of multiphoton transitions [33].

The analysis shows that for the $\Lambda$-system in some directions around second order traps the objective increases. Such behavior was also found for a four-level system by constructing a second order trap where the objective increases in a direction $\delta e(t) = \chi_{[0,\pi]}(t)$ [35] (Example 2). However, this increase is slow (at best it is of 4th order in $\delta e$ in our case, and of 3rd order in the example of [35]) and simple gradient methods may not be effective in escaping these critical points. Therefore more sophisticated algorithms exploiting the higher order information about the objective are generally necessary to climb up the landscape in the vicinity of second order traps.

### 3 Numerical analysis

In this section we consider piecewise constant controls of the form

$$
\varepsilon(t) = \sum_{k=1}^M c_k \chi_{[t_k,t_{k+1}]}(t)
$$

where $\chi_{[t_k,t_{k+1}]}(t)$ is the characteristic function of the interval $[t_k,t_{k+1}]$ (i.e., $\chi_{[t_k,t_{k+1}]}(t) = 1$ if $t \in [t_k,t_{k+1}]$ and $\chi_{[t_k,t_{k+1}]}(t) = 0$ otherwise), $t_k = \Delta t(k-1)$ and $\Delta t = T/M$. In this representation the control is an $M$-dimensional vector $C = (c_1,\ldots,c_M)$. The objective is defined as

$$
J(C) = \text{Tr}[U_T \rho_0 U_T^\dagger O], \quad \text{where } U_T = U_T(C)
$$

The gradient of the objective is

$$
\nabla J(C) = \left( \frac{\partial J(C)}{\partial c_1}, \ldots, \frac{\partial J(C)}{\partial c_M} \right)
$$
Here the partial derivative with respect to $c_l$ has the form

$$\frac{\partial J(C)}{\partial c_l} = 2\Delta t \cdot \text{Im} \left( \text{Tr} \left[ W_l^\dagger V W_l \rho_0 U_l^\dagger O U_T \right] \right)$$

where

$$W_l = U_l U_{l-1} \ldots U_2 U_1, \quad U_k = e^{-i(H_0 + c_k V)\Delta t}, \quad U_T = W_M$$

We use GRAPE [15] and BFGS from the MATLAB Optimization Toolbox as local search algorithms. The GRAPE algorithm starts with a randomly generated initial control $C_1$, computes the gradient $\nabla J(C_1)$, updates the control as $C_2 = C_1 + \epsilon \nabla J(C_1)$, where $\epsilon > 0$ is a small number, and continues the loop either until the objective reaches the value $J_{\text{stop}} = 1 - I_{\text{err}}$, where $I_{\text{err}}$ is some threshold or until some number $K_{\text{stop}}$ of iterations is reached. The components of the initial control $C_1$ are generated randomly with uniform distribution in the interval $[-c_0, c_0]$ with some $c_0 > 0$. Since the second order trap is at $\varepsilon(t) = 0$, $c_0$ can be viewed as measure of distance from the second order trap.

We consider the following parameters of the $\Lambda$-system:

$$H_0 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2.5 \end{pmatrix}, \quad V = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 1.7 \\ 1 & 1.7 & 0 \end{pmatrix}$$

The final time in the simulations is chosen as $T = 10$, that is several times larger than typical oscillation periods in the system. First we consider the case $\lambda = 0$ which corresponds to the standard $P_{1\to2}$ problem, where the objective is to transfer the population from the ground state $|1\rangle$ to the intermediate state $|2\rangle$. For this case, second order traps were not discovered in [30]. Figure 2 shows that for this case it is easy to find, even in a small number of iterations, control fields that produce an objective which deviates from the global maximum by as little as $10^{-5}$. Figure 3(left) shows the distribution of the number of iterations required to achieve 99.999% population transfer and (right) the distribution of initial objective values. It is seen that despite the fact that the search generally starts from low objective values, i.e. the bottom of the landscape, it always reaches the top in a small number of iterations.

Now we consider the case $\lambda > 0$, with the goal to analyze how for small initial controls the $\varepsilon(t) = 0$ second order trap may significantly slow down the search for global maxima. Recall that $O = |2\rangle\langle 2| - \lambda |3\rangle\langle 3|$, and therefore larger values of $\lambda$ should have a more significant effect on the slowdown of the search. We choose $\lambda = 5$ and perform the optimization for $c_0 = 0.1, 0.2, 0.3, \ldots, 1$. For each choice of $c_0$, $L = 100$ runs of the GRAPE algorithm were performed with components of the initial controls generated randomly in the interval $[-c_0, c_0]$. Other parameters were chosen as follows: $K_{\text{stop}} = 1000$ for the maximum allowed number of iterations, $I_{\text{err}} = 0.1$ for the allowed deviation from the global maximum of the objective, $\epsilon = 0.1$ for the iteration size, $T = 10$ for the final time, and $M = 200$ for the number of components in each control.

For each $c_0$ we determine the number of unsuccessful attempts $N_{\text{fail}}$ (among $L$ runs) when the algorithm fails to reach the objective value $1 - I_{\text{err}}$ in less than $K_{\text{stop}}$ iterations. We also determine the mean number of iterations required to reach the objective value $1 - I_{\text{err}}$ in the successful $L - N_{\text{fail}}$ runs. These data are plotted in Fig. 4. For $c_0 = 0.1$ and $c_0 = 0.2$ we find that $N_{\text{fail}} = L$, showing that all runs of the algorithm fail if the amplitudes $c_i$ of
Figure 2: (Color online) A typical example of the behavior of the objective and the norm of the gradient vs number of iterations for maximizing the $P_{1\rightarrow 2}$ transition probability for a three-level Λ-system. The parameters are: $M = 200$, $c_0 = 1$, $\epsilon = 0.1$, $I_{\text{err}} = 10^{-5}$. The objective value 0.99999 is reached in 323 iterations.

Figure 3: (Color online) $L = 1000$ initial controls were randomly generated to produce these plots for maximizing $P_{1\rightarrow 2}$ in a three-level Λ-system ($M = 200$, $c_0 = 1$, $\epsilon = 0.1$, $I_{\text{err}} = 10^{-5}$). Left histogram represents the distribution of the number of iterations required to reach the objective value $J = 1 - I_{\text{err}} = 0.99999$ (minimum number of iterations is 165, maximum 1400, mean 388, and the standard deviation $\sigma = 125$). Right histogram represents the distribution of the initial objective values. In each histogram 25 intervals were used.
the initial control satisfy \( c_i \leq 0.2 \ (i = 1, \ldots, M) \). The number of failed runs decreases with increase of \( c_0 \) and becomes close to zero when \( c_0 = 1 \). Starting from \( c_0 = 0.3 \), the mean number of iterations required to reach the desired objective value decreases linearly. We also observe that if the limiting number of iterations \( K_{\text{stop}} \) is increased then some runs will be successful even for small \( c_0 \). However, in practice there always exist limits on the number of possible iterations and therefore if a large number of iterations is required to reach a desired objective value, for practical purposes this may look like a trap. Another comment is that the number of iterations might be dependent on the details of the algorithm. For example, using an adaptive step size may improve performance of the gradient search at points with small gradient. For comparison we use the BFGS method. The number of fails for every \( c_0 \) is less than for GRAPE, particularly for small \( c_0 \), as plotted by the dashed line in Fig. [4]. The decrease in the efficiency of the linear gradient search was also observed for quantum systems which have forbidden transitions between distant energy levels [34]. The three-level \( \Lambda \)-system has a forbidden 1 \( \rightarrow \) 2 transition and therefore is within this class. Our work however studies a different dependence of the efficiency — on \( c_0 \), which is effectively the distance of the initial control from the second order trap.

**Conclusions**

This work shows that constant controls, which are second order traps in the control landscape for a three-level \( \Lambda \)-system, are not local maxima: there exists directions around these controls in which the objective increases at 4th order. A numerical investigation shows that indeed the search with simple gradient methods becomes slow in a sufficiently small neighborhood of \( \varepsilon(t) = 0 \). These results suggest that in the vicinity of second order traps, simple gradient algorithms may not be sufficient, and more sophisticated algorithms that exploit higher order derivative information may be necessary.

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Figure 4: (Color online) For each point, $L = 100$ runs of either GRAPE (with $\epsilon = 0.1$) or BFGS were performed with initial controls generated randomly in the interval $[-c_0, c_0]$ ($T = 10$, $M = 200$, and $L_{\text{err}} = 0.1$). The bottom two (blue) lines show the dependence of the number of failed runs for GRAPE (solid line, using $K_{\text{stop}} = 1000$) and BFGS (dashed line, with the termination condition determined by the default MATLAB criterion) plotted with a fourth order polynomial interpolations. Upper (green) line shows the mean number of iterations for those runs of GRAPE that were successful, i.e. that found controls that achieve the fidelity 0.9 in fewer than $K_{\text{stop}} = 1000$ iterations (linear interpolation).
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