Are there Traps in Quantum Control Landscapes?

Alexander N. Pechen and David J. Tannor

Phys. Rev. Lett. **106**, 120402 — Published 22 March 2011

DOI: 10.1103/PhysRevLett.106.120402

LB12609

Are there traps in quantum control landscapes?

Alexander N. Pechen and David J. Tannor

Weizmann Institute of Science, Rehovot, Israel

There has been great interest in recent years in quantum control landscapes. Given an objective J that depends on a control field ε the dynamical landscape is defined by the properties of the Hessian $\delta^2 J/\delta\varepsilon^2$ at the critical points $\delta J/\delta\varepsilon = 0$. We show that contrary to recent claims in the literature the dynamical control landscape can exhibit trapping behavior due to the existence of special critical points and illustrate this finding with an example of a 3-level Λ-system. This observation can have profound implications for both theoretical and experimental quantum control studies.

PACS numbers: 02.30.Yy, 32.80.Qk

Quantum control aims to manipulate the dynamics of physical processes on the atomic and molecular scale. It is a rapidly growing field of science with numerous applications ranging from selective laser-induced atomic or molecular excitations to high harmonic generation, quantum computing and quantum information, and control of chemical reactions by specially tailored laser pulses, etc. [1–5].

Generally quantum control problems can be formulated as the maximization of an objective function $J(\varepsilon)$ by a suitable optimal control ε. A wide variety of quantum control phenomena, selective bond breaking, etc. can be described by control objectives of the form $J(\varepsilon) = \mathrm{Tr}[U_\varepsilon(T)\rho_0 U_\varepsilon^\dagger(T) O]$, where O is an operator describing the target, ρ_0 is the initial density matrix and $U_\varepsilon(T)$ is the evolution operator under the action of the control ε satisfying the equation

$$\frac{dU_\varepsilon(t)}{dt} = -i[H_0 - \mu\varepsilon(t)]U_\varepsilon(t)\,, \tag{1}$$

where H_0 is the free system Hamiltonian and μ is the dipole moment.

The objective $J = J[\varepsilon]$ as a function of the control ε defines the landscape of the control problem. The structure of the landscape determines the complexity of the underlying control problem. Particularly important features of a control landscape are traps — local maxima of $J(\varepsilon)$. Traps can have a profound influence on both theoretical and experimental quantum control studies — they can slow down or even prevent finding globally optimal controls and can lead to erroneous physical conclusions about optimal processes and robustness. We show that contrary to recent claims in the literature [6–11] the dynamical control landscape can exhibit trapping behavior due to the existence of special critical points and illustrate this finding with an example of a 3-level Λ-system. This observation can have profound implications for both theoretical and experimental quantum control studies.

To understand why traps are significant, consider the generic problem of finding a globally optimal control ε_* such that $J(\varepsilon_*) = J_{\max} = \max_\varepsilon J(\varepsilon)$. Unless the system is extremely simple, numerical or laboratory optimization algorithms generally need to be employed. The prevailing theoretical methods start from an initial trial control ε and use gradient and Hessian (first- and second-order) information to explore the neighborhood for a control with better performance. This new control is then used as a new starting point and the process is iterated. Experimentally, evolutionary algorithms are commonly used. While these algorithms are not strictly first- or second-order, each new generation of controls still has a propensity to explore the neighborhood of the previous generation. If the control

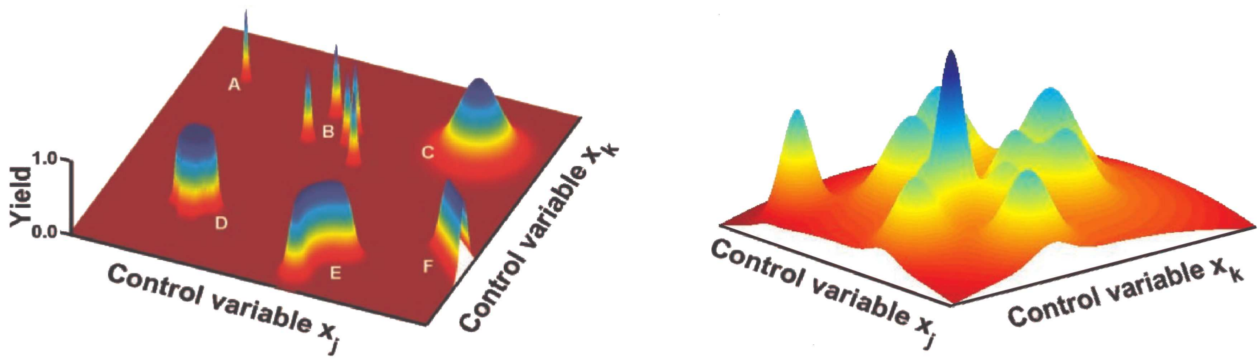

FIG. 1. (Color online). Left: Cartoon of a landscape without traps (local maxima). All peaks are of the same height and thus all of them are global maxima. From *Science* **303**, 1998 (2004). Reprinted with permission from AAAS. Right: Cartoon of a landscape with traps. The landscape has one highest peak representing the global maximum and several peaks of lower height representing multiple local maxima. Both landscapes are plotted for two control variables, x_j and x_k, representing the control field $\varepsilon(t)$ at two different time moments. The actual number of variables in practical applications may be several hundreds. A local search over the landscape on the left will eventually reach a global maximum, due to the absence of traps. However, a local search over the landscape on the right will most likely find a trap, ending the search process without ever finding the highest peak.

landscape has traps then first- and second-order algorithms, which effectively are providing only a local search over this landscape, can be prevented from reaching a globally optimal solution ε_*. Thus, the existence or absence of traps is a significant characteristic for any control landscape. Fig. 1 shows landscapes with and without traps.

The analysis of quantum control landscapes was performed in a series of pioneering works [6–11]. Extrema of trace functions over unitary and orthogonal groups were also studied in a different context [12, 13] and in the context of quantum control [14, 15]. The analysis in [6–11] concluded the absence of traps. In subsequent work it was established that this conclusion was under the implicit assumption that the Jacobian $\delta U_\varepsilon/\delta\varepsilon$ has full rank at any point. Although this assumption was shown to be violated at times [20], is was believed to be generally applicable. Recently, a particular example of a trap was constructed [21]. The present paper significantly advances the field by showing that second order traps — points at which the Hessian $H = \delta^2 J/\delta\varepsilon$ is negative semi-definite — exist in a wide class of quantum control systems [22].

We begin our discussion by distinguishing between the dynamic and the kinematic control landscapes. Until now we have been discussing the functional $J[\varepsilon]$, but one may also consider the simpler functional $J[U]$, where the dependence of U on ε is suppressed:

$$J_{\rm K}[U] = {\rm Tr}[U\rho_0 U^\dagger O] \tag{2}$$

Equation (2) defines the kinematic control landscape. A dynamic critical point (DCP) is defined by $\nabla J(\varepsilon) = \delta J(\varepsilon)/\delta\varepsilon = 0$ whereas a kinematic critical point (KCP) is defined by $\nabla J_{\rm K}(U) = \delta J(U)/\delta U = 0$, where ∇ denotes gradient. Dynamic and kinematic traps are subopimal maxima for $J[\varepsilon]$ and $J_{\rm K}[U]$, respectively.

Assuming complete controllability, i.e. that U in (2) can be any unitary operator, the *kinematic* control landscape is known to be free of traps: all critical points of $J_{\rm K}[U]$ are either global maxima and minima, or saddles [16]. This result implies that the *dynamic* control landscape will be trap-free if one additionally assumes that the Jacobian $\delta U_\varepsilon(T)/\delta\varepsilon$ has full rank at any ε [17, 18]. Indeed, by the chain rule,

$$\frac{\delta J(\varepsilon)}{\delta\varepsilon} = \frac{\delta J_{\rm K}[U_\varepsilon(T)]}{\delta U_\varepsilon(T)}\frac{\delta U_\varepsilon(T)}{\delta\varepsilon}$$

and hence under the full rank condition all DCP are at KCP and have exactly the same critical point structure as the corresponding KCP [19].

Our first result concerns the inequivalence of critical point structures. To find the condition for a KCP of (2) we take any infinitesimal variation of U in the form $U \to U' = U(1+\delta U)$ [16]. Unitarity of U' up to the first order in δU implies $\delta U^\dagger = -\delta U$, i.e. δU is anti-Hermitian, and hence the variation of the objective $J_{\rm K}$ with respect to U is $\delta J_{\rm K} = J_{\rm K}[U'] - J_{\rm K}[U] = {\rm Tr}\{\delta U[\rho_0, O_T]\} + o(\|\delta U\|)$, where $O_T = U^\dagger O U$. If U is a critical point for $J_{\rm K}[U]$, then the condition $\delta J_{\rm K} = 0$ needs to hold for any anti-Hermitian δU, implying that

$$[\rho_0, O_T] = 0\,. \tag{3}$$

Equation (3) is the condition for a KCP. All U satisfying the condition (3) were shown to be either global maxima, minima, or saddles [16], and therefore second order traps do not exist for kinematic control landscapes. Dynamical critical control fields that violate (3) were shown to exist for the problem of optimal population transfer between two pure states of a quantum systems [21]. We have been able to generalize this finding by showing that such fields exist not only for optimal population transfer between two pure states, but for maximizing the expectation value of a more general class of observables. The proof is given in Section 3 of the Supplementary material [23]. This inequivalence of the critical points in the dynamic and kinematic landscapes is an indication of the breakdown of the full rank assumption.

We now turn to our main result. For a general class of systems there exist dynamical critical controls ($[\rho_0, O_T] = 0$) that are second order traps due to the violation of the full rank assumption. In particular, second order traps appear in the dynamical control landscape whenever the dipole moment satisfies $\mu_{ij} = 0$ for some $i \neq j$ (i.e. if a direct transition between some pair of levels is forbidden). In this case there exists an initial density matrix and a target operator such that $\varepsilon(t) = 0$ is a second order trap. (Note that the condition $\mu_{ij} = 0$ can be consistent with the assumption of complete controllability of the system provided that the levels i and j are connected indirectly through other states.) More generally, a control $\varepsilon(t) = \varepsilon_0$ is a second order dynamical trap if in terms of the spectral decomposition $\tilde{H}_0 = H_0 - \mu\varepsilon_0 = \sum_{i=1}^{n} \tilde{h}_i |\tilde{i}\rangle\langle\tilde{i}|$ the initial density matrix and target operator have the form $\rho_0 = |\tilde{k}\rangle\langle\tilde{k}|$ and $O = \sum_{i=1}^{n} \lambda_i |\tilde{i}\rangle\langle\tilde{i}|$, 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 $i < k$. (Again, the condition that $\langle\tilde{i}|\mu|\tilde{k}\rangle = 0$ for any $i < k < n$ can be consistent with the controllability assumption if the dipole moment connects the state $|\tilde{k}\rangle$ with all states $|\tilde{i}\rangle$ for $i < k$ through other states.) To prove this finding, we explicitly compute the Hessian and show that it is negative semidefinite under the above conditions; the details of the proof are given in Section 2 of the Supplementary material [23].

The simplest example of such a second order trap appears in the problem of maximizing the expectation of an operator $O = \sum_{i=1}^{3} \lambda_i |i\rangle\langle i|$ with $\lambda_2 > \lambda_1 > \lambda_3$ for a three-level Λ-atom initially in the state $\rho_0 = |1\rangle\langle 1|$. The dipole moment for Λ-atom satisfies $\mu_{12} = 0$, consistent with the controllability assumption if $\mu_{13} \neq 0$ and $\mu_{23} \neq 0$. Globally optimal control fields steer $|1\rangle$ completely into $|2\rangle$ producing the global maximum of the objective with value $J_{\max} = \lambda_2$. The control field $\varepsilon(t) = 0$ produces a second order trap with the objective value $J = \lambda_1 < J_{\max}$.

In conclusion, we have established that second order traps in quantum control landscapes exist in a wide range of quantum systems. More research will be required to establish if these points are true traps, but for the local search algorithms currently in use second order traps

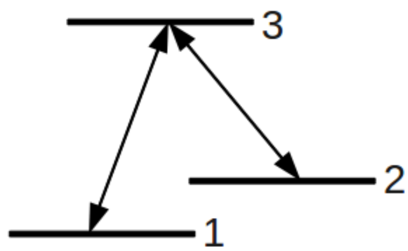

FIG. 2. The simplest example of a quantum system possessing a second order trap is a 3-level Λ-system initially in the ground state. The control field $\varepsilon(t) = 0$ is a second order trap for maximizing expectation of any target operator of the form $O = \sum_{i=1}^{3} \lambda_i |i\rangle\langle i|$ with $\lambda_2 > \lambda_1 > \lambda_3$.

pose virtually all the same numerical and experimental difficulties as true traps. Moreover, since the present work establishes that the full rank assumption is violated for a wide class of quantum systems, the previous claims of the absence of traps, which were based on this assumption, have to be completely rethought.

This research is made possible in part by the historic generosity of the Harold Perlman family. A. Pechen acknowledges partial support from the EMALI European Union Marie Curie Teaching-Research Network Contract No. MRTN-CT-2006-035369.

[1] P. W. Brumer and M. Shapiro, *Principles of the Quantum Control of Molecular Processes*, (Wiley-Interscience, 2003).

[2] D.-J. Tannor, *Introduction to Quantum Mechanics: A Time Dependent Perspective*, (University Science Press, Sausalito, 2007).

[3] V. S. Letokhov, *Laser Control of Atoms and Molecules*, (Oxford University Press, USA, 2007).

[4] C. Brif, R. Chakrabarti, and H. Rabitz, New J. Phys. **12**, 075008 (2010).

[5] H. M. Wiseman and G. J. Milburn, *Quantum Measurement and Control*, (Cambridge University Press, Cambridge, 2010).

[6] H. Rabitz, M. Hsieh, and C. Rosenthal, Science **303**, 1998 (2004).

[7] H. Rabitz, J. Modern Optics **51**, 2469 (2004).

[8] H. Rabitz, M. Hsieh, and C. Rosenthal, Phys. Rev. A **72**, 052337 (2005).

[9] A. Rothman, T.-S. Ho, and H. Rabitz, Phys. Rev. A **73**, 053401 (2006).

[10] M. Hsieh, R. Wu, C. Rosenthal, and H. Rabitz, J. Phys. B: At. Mol. Opt. Phys. **41**, 074020

(2008).

[11] M. Hsieh and H. Rabitz, Phys. Rev. A **77**, 042306 (2008).

[12] J. von Neumann, Tomsk Univ. Rev. **1**, 286 (1937).

[13] R. Brockett, Lin. Alg. Appl. **122/123/124**, 761 (1989).

[14] S. J. Glaser, *et al.*, Science **280**, 421 (1998).

[15] M. D. Girardeu, S. G. Schirmer, J. V. Leahy, and R. M. Koch, Phys. Rev. A **58**, 2684 (1998).

[16] H. Rabitz, M. Hsieh, and C. Rosenthal, J. Chem. Phys. **124**, 204107 (2006).

[17] T.-S. Ho and H. Rabitz, J. Photochemistry and Photobiology A: Chemistry **180**, 226 (2006).

[18] R. Chakrabarti and H. Rabitz, Intl. Rev. Phys. Chem. **26**, 671 (2007).

[19] R. Wu, A. Pechen, H. Rabitz, M. Hsieh, and B. Tsou, J. Math. Phys. **49**, 022108 (2008).

[20] R. Wu, J. Dominy, T.-S. Ho, and H. Rabitz, *Singularities of quantum control landscapes*, Preprint arXiv:0907.2354.

[21] P. de Fouquieres and S. G. Schirmer, *Quantum control landscapes: a closer look*, Preprint arXiv:1004.3492.

[22] Second order traps, where the Hessian H_ε is negative semi-definite, are not necessarily sub-optimal maxima: if the kernel of H_ε is non-empty and the third or higher order variations of J are non-zero along a $\delta\varepsilon \in \ker H_\varepsilon$, then ε may be neither a local maximum nor minimum. They are however important as being traps for commonly used algorithms exploiting only the first and second order information.

[23] See supplementary material.