Topology of optimally controlled quantum mechanical transition probability landscapes

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(Received 29 September 2005; published 27 July 2006)

An optimally controlled quantum system possesses a search landscape defined by the physical objective as a functional of the control field. This paper particularly explores the topological structure of quantum mechanical transition probability landscapes. The quantum system is assumed to be controllable and the analysis is based on the Euler-Lagrange variational equations derived from a cost function only requiring extremizing the transition probability. It is shown that the latter variational equations are automatically satisfied as a mathematical identity for control fields that either produce transition probabilities of zero or unit value. Similarly, the variational equations are shown to be inconsistent (i.e., they have no solution) for any control field that produces a transition probability different from either of these two extreme values. An upper bound is shown to exist on the norm of the functional derivative of the transition probability with respect to the control field anywhere over the landscape. The trace of the Hessian, evaluated for a control field producing a transition probability of a unit value, is shown to be bounded from below. Furthermore, the Hessian at a transition probability of unit value is shown to have an extensive null space and only a finite number of negative eigenvalues. Collectively, these findings show that (a) the transition probability landscape extrema consists of values corresponding to no control or full control, (b) approaching full control involves climbing a gentle slope with no false traps in the control space and (c) an inherent degree of robustness exists around any full control solution. Although full controllability may not exist in some applications, the analysis provides a basis to understand the evident ease of finding controls that produce excellent yields in simulations and in the laboratory.

DOI: 10.1103/PhysRevA.74.012721 PACS number(s): 03.65.Nk, 03.67.Lx

I. INTRODUCTION

The control of quantum phenomena [1–3], especially with tailored laser pulses, is an active area of theoretical [4–21] and experimental research [22–29]. A common foundation of much quantum control research is optimal control theory (OCT) formulated within quantum dynamics [4–7] and the realization of optimal control experiments (OCEs) [22–29] in conjunction with closed-loop learning control techniques [8]. Despite the myopic nature of the local search algorithms commonly employed in typical quantum OCT calculations, excellent target yields are frequently achieved in the numerical simulations. More importantly, in spite of the curse of dimensionality arising from searching through the usually very high dimensional space of control variables corresponding to pulse shaper phases and amplitudes in typical OCE studies, successful control results can usually be determined quickly in the laboratory. This paper aims to explain why it appears possible to consistently find successful controls with evident ease. The key to this behavior lies in the topology of the quantum control landscapes, defined as a physical objective as a functional of the control laser field.

Although many particular physical objectives have been considered in control studies, a common goal is the maximization of the probability $P_{i\to f}$ of making a transition from initial state $|i\rangle$ to final state $|f\rangle$. These states are typically eigenstates of the field free Hamiltonian H_0 where the full Hamiltonian often has the form $H=H_0-\mu\varepsilon(t)$ with μ being the dipole moment operator and $\varepsilon(t)$ being the control electric field. A suitable temporal shape for $\varepsilon(t)$ is sought that maximizes $P_{i\to f}$ at target time T, which may be finite or asymptotic with $T\to\infty$. In practice, other ancillary costs may also be imposed, including on the form of the laser pulse or minimization of its energy. Nevertheless, the fundamental control problem for analysis is

$$\operatorname*{Max}_{\varepsilon(t)} P_{i\to f}, \tag{1}$$

and this paper is concerned with analyzing the structure of the control landscape

$$P_{i\to f}=P_{i\to f}[\varepsilon(t)], \tag{2}$$

which is a functional of the control field [30]. In the laboratory other factors can enter, but an ultimate common goal is the clean performance of state-to-state transfer in Eq. (1). Knowledge of the general topology of this landscape, including its extrema values, slopes, and curvature, is fundamental to understanding the ability of finding good quality robust controls in the laboratory and in simulations.

This paper will carry out an analysis of the control landscape in Eq. (2), treating $\varepsilon(t)$ as an arbitrary continuous temporal function. As a result, the search to maximize $P_{i\to f}$ is formally over an infinite dimensional space. However, in practical simulations or in the laboratory, the control field is always discretized in some fashion in either the time or the frequency domain. The number of discretized control variables can still remain very high (e.g., hundreds of phase and amplitude frequency domain controls are often employed in the laboratory [22–29]). Thus, regardless of the representation used for the control, the maximization of $P_{i\to f}$ generally entails a search through a high dimensional control space for an optimal field. With no further information available, the natural expectation is that the control landscape in Eq. (2) would likely have a highly complex topology with perhaps many maxima, minima, and saddle points. In particular, it is reasonable to expect that suboptimal local maxima would be encountered with values $P_{i\to f}<1.0$.

The present analysis will assume that the quantum system is controllable [31–34] such that in principle one can steer the transition probability about at will. However, this statement in itself does not evidently preclude the possibility of local maxima existing to act as traps, especially for control searches employing local algorithms (e.g., gradient methods), which would naturally stop at the first local maximum. The many successful control simulations producing high yield outcomes [4–21] for $P_{i\to f}$ over a variety of chemical and physical applications as well as the increasing number of successful closed-loop adaptive control experiments [22–29] suggest that the landscape in Eq. (2) has a very favorable topology regardless of the detailed nature of the Hamiltonian. Furthermore, the success of the rapidly increasing number of experiments also suggest that there is an inherent degree of robustness at the landscape extrema to the inevitable presence of laboratory noise.

This paper will analyze the topology of the landscape in Eq. (2) to reveal the origins of the observations above. More complex circumstances can arise, including systems in mixed states and the presence of decoherence processes. Nevertheless, the analysis here provides the basis for assessing the basic features of controlling quantum phenomena. An initial effort along these lines was undertaken based on starting with the formulation of the system unitary evolution operator expressed in the form $U(T)=\exp(iA)$ where $A^\dagger=A$ [30,35]. It was argued that the control variables may be transferred from $\varepsilon(t)$ to effectively being the elements of the action matrix A. This abstraction revealed valuable information about the control landscape, although it left wanting an explicit connection to the control field $\varepsilon(t)$, which is naturally the true function being varied either in simulations or in the laboratory. Thus, the present paper will explicitly work with the control field $\varepsilon(t)$ to make the landscape analysis transparent in a physical context. A formal Lie group analysis of $P_{i\to f}$ was also undertaken [36,37], and the mathematical underpinnings of this analysis can be traced back to work in the early development period of quantum mechanics [38]. Finally, an earlier Hessian (stability) analysis of $P_{i\to f}$ was carried out with other costs involved besides that of Eq. (1), but the complexity of the resultant situation gave only a partial glimpse of the fundamental underlying landscape [39]. The present work will go beyond these prior studies to reveal the landscape topology in the familiar physical context of the control field $\varepsilon(t)$ and employing the standard variation formulation based on Eq. (1).

Section II will present the variational formulation along with first order (gradient) and second order Hessian analyses. Special emphasis will be given to (a) identifying the conditions under which the variational equations are identically satisfied or violated, as well as (b) seeking bounds on the slope and curvature over the control landscape. Section III will present concluding remarks on the physical significance of the findings in this paper.

II. OPTIMAL CONTROL VARIATIONAL EQUATIONS AND THEIR ANALYSIS

The quantum system subjected to control in this work is assumed to be of arbitrary, but finite dimension N. Nominally this would exclude controlled molecular dissociation where a continuum of final states is involved, however in practice, one may model such problems as having a finite (possibly large) number of levels [40,41]. The system is assumed to have the common Hamiltonian structure $H(t)=H_0-\mu\varepsilon(t)$, although other forms could just as well be treated. Importantly, the system is assumed to be controllable such that, in principle, at least one field exists that will permit an exact transition from an arbitrary initial state $|i\rangle$ to the final state $|f\rangle$. A formal set of analysis tools is available to assess controllability starting with knowledge of the operators H_0 and μ [31–34]. In practice, testing for controllability can be a tedious process, and it has only been established in a limited number of systems. A statistical assessment starting from a related connectivity analysis perspective suggests that only a null set of quantum systems is likely to be uncontrollable [42]. These analyses, along with the empirical evidence from the many high quality numerical optimal control calculations [4–21] and now the emerging experiments even on complex systems [22–29], suggests that most realistic applications involve systems that are either fully controllable or acceptably so from a practical perspective. The presence of a high density of states *per se* does not imply a lack of controllability, although bandwidth or other restrictions on the applied controls may in practice limit the achieved outcome to being less than ideal. The present work will accept the premise that the quantum system is controllable, and this point will be exploited in the analysis below.

The control cost function is taken to have the following standard form [7]:

$$J=\langle i|U^\dagger(T,0)|f\rangle\langle f|U(T,0)|i\rangle + \mathrm{Im}\int_0^T \langle\lambda(t)|\left(\imath\hbar\frac{\partial}{\partial t}-H_0+\mu\varepsilon(t)\right)|\psi(t)\rangle dt, \qquad (3)$$

where $|\lambda(t)\rangle$ is the Lagrange multiplier state introduced to preserve satisfaction of Schrödinger's equation and $U(T,0)$ is the system time evolution operator evaluated at the target time T. As the last term in Eq. (3) is zero upon satisfaction of Schrödinger's equation for the evolving quantum state $\psi(t)$, it is evident that J takes on the value $P_{i\to f}=|\langle f|U(T,0)|i\rangle|^2$. The three Euler-Lagrange equations associated with demanding that $\delta J=0$ are

$$\imath\hbar\frac{\partial}{\partial t}|\psi(t)\rangle=[H_0-\mu\varepsilon(t)]|\psi(t)\rangle, \quad |\psi(0)\rangle=|i\rangle, \qquad (4)$$

$$\imath\hbar\frac{\partial}{\partial t}|\lambda(t)\rangle = [H_0 - \mu\varepsilon(t)]|\lambda(t)\rangle,$$

$$|\lambda(T)\rangle = -\frac{2}{\hbar}|f\rangle\langle f|U(T,0)|i\rangle, \quad (5)$$

$$\frac{\delta J}{\delta\varepsilon(t)} = \text{Im}[\langle\lambda(t)|\mu|\psi(t)\rangle]$$
$$= \frac{2}{\hbar}\text{Im}[\langle i|U^\dagger(t,0)\mu U(t,0)U^\dagger(T,0)|f\rangle\langle f|U(T,0)|i\rangle]$$
$$= \frac{2}{\hbar}\text{Im}[\langle i|\mu(t)|q\rangle] = 0, \quad (6)$$

where

$$\mu(t) = U^\dagger(t,0)\mu U(t,0) \quad (7)$$

and

$$|q\rangle = U^\dagger(T,0)|f\rangle\langle f|U(T,0)|i\rangle. \quad (8)$$

The distinct but equivalent forms of $\delta J/\delta\varepsilon(t)$ in Eqs. (6) will be exploited in different particular circumstances below. In Eqs. (7) and (8), $\mu(t)$ is the evolution of the system dipole under the full controlled dynamics and $|q\rangle$ is a state, which under the system dynamics would generally be a superposition of the eigenstates of H_0. The remainder of this section is concerned with the conditions under which $\delta J/\delta\varepsilon(t)=0,\ \forall t$, as well as with other associated behavior of J.

A. When can $\delta J/\delta\varepsilon(t)=0,\ \forall t$ be satisfied?

This section will consider three circumstances where control fields will either satisfy or not satisfy the right-hand side of Eq. (6) being zero. Naturally the cases of $P_{i\to f}=0$ and 1.0 are extrema of the landscape, and it is expected that the variational equations are exactly satisfied for these cases. This conclusion will be explicitly confirmed in Secs. II A 1 and II A 2, followed by an analysis of the circumstance $0<P_{i\to f}<1.0$.

1. Null control conditions $P_{i\to f}=0$

The situation considered here either corresponds to the introduction of no control field, or where a control field is present but we have $\langle f|U(T,0)|i\rangle=0$. The case of no control corresponds to $\langle f|U(T,0)|i\rangle=\langle f|i\rangle\exp(-\imath E_iT/\hbar)=0$, where E_i is the eigenvalue associated with the ith state of H_0 and the final state $|f\rangle$ is naturally assumed to have no overlap with the initial state $|i\rangle$. In the case where a control is present under these circumstances, there will be evolution $|\psi(t)\rangle=U(t,0)|i\rangle$ from one point in the landscape of Eq. (2), where $|\psi(0)\rangle=|i\rangle$ to another point in the landscape, where $|\psi(T)\rangle=U(T,0)|i\rangle$, such that $\langle f|\psi(T)\rangle=0$. The path between these two landscape points can be highly complex, and the analysis of such evolution is the subject of Sec. II A 3 below. It is evident from Eq. (8) that $|q\rangle=0$ under both circumstances above. Thus, we have that $\delta J/\delta\varepsilon(t)=0$, $\forall t$ for those controls satisfying $P_{i\to f}=0$. From a previous analysis on the multiplicity of control solutions [43,44], we can generally expect an infinite number of nontrivial control fields to exist, carrying out the mapping $|i\rangle\to U(T,0)|i\rangle$, where $\langle f|U(T,0)|i\rangle=0$, from one location to another in the control landscape, all corresponding to extreme minima points where $P_{i\to f}=0$. Thus, in cases of either no control field or when the control takes the system from one null transition probability value to another in the landscape, the Euler-Lagrange equations are automatically satisfied.

2. Perfect control conditions $P_{i\to f}=1.0$

This case corresponds to the presence of a control field $\varepsilon(t)$ introducing dynamics such that

$$U(T,0)|i\rangle = e^{\imath\phi}|f\rangle, \quad (9)$$

with ϕ being an arbitrary real phase. In this circumstance, there is perfect control with $P_{i\to f}=|\langle f|U(T,0)|i\rangle|^2=1.0$. By operating on the expression in Eq. (9) with $U^\dagger(T,0)$ it is easy to show that $U^\dagger(T,0)|f\rangle=e^{-\imath\phi}|i\rangle$, implying that $U^\dagger(T,0)|f\rangle\langle f|U(T,0)=|i\rangle\langle i|$. Substituting the latter relation into Eq. (8) leads to $|q\rangle=|i\rangle$ which reduces Eq. (6) to

$$\frac{\delta J}{\delta\varepsilon(t)} = \frac{2}{\hbar}\text{Im}[\langle i|\mu(t)|i\rangle] = 0. \quad (10)$$

The latter equality follows from $\mu(t)$ being a Hermitian operator with a real expectation value. We may conclude that control fields producing dynamics leading to $P_{i\to f}=1.0$ will automatically satisfy the variational Eqs. (4)–(6) including $\delta J/\delta\varepsilon(t)=0,\ \forall t$. The multiplicity of control solutions [43,44] implies that the landscape will have many, if not an infinite number, of such perfect control extrema, each accessed by a unique control field $\varepsilon(t)$.

3. Behavior of $\delta J/\delta\varepsilon(t)$ for arbitrary control fields

The analysis in Secs. II A 1 and II A 2 above formally confirms that the control landscape has critical points corresponding to either no control or perfect control satisfying the variational equations. The remaining question is whether other control fields $\varepsilon(t)$ might exist satisfying the Euler-Lagrange variational conditions, Eqs. (4)–(6), producing $0<P_{i\to f}<1.0$. Equations (4) and (5) by construction will always be satisfied by an arbitrary control field $\varepsilon(t)$. Thus, the question reduces to whether Eq. (6) can be satisfied such that $\delta J/\delta\varepsilon(t)=0,\ \forall t$. Satisfaction of the latter relation also implies that the nth derivative of $\delta J/\delta\varepsilon(t)$ must in turn satisfy the relation

$$\frac{\partial^n}{\partial t^n}\left(\frac{\delta J}{\delta\varepsilon(t)}\right) = 0, \quad \forall\, t, \quad n=1,2,\ldots\ . \quad (11)$$

These derivatives may be readily evaluated from Eqs. (6) and (7) using the relations

$$\frac{d\mu(t)}{dt} = \left(\frac{-\imath}{\hbar}\right)[\mu(t),H(t)], \quad (12)$$

$$\frac{dH(t)}{dt} = -\frac{d\varepsilon(t)}{dt}\mu(t). \tag{13}$$

In particular, the first and second derivatives in Eq. (11) are

$$\frac{\partial}{\partial t}\frac{\delta J}{\delta\varepsilon(t)} = \frac{2}{\hbar}\,\mathrm{Im}\left[\left(\frac{-\imath}{\hbar}\right)\langle i|[\mu(t),H(t)]|q\rangle\right], \tag{14}$$

$$\frac{\partial^2}{\partial t^2}\frac{\delta J}{\delta\varepsilon(t)} = \frac{2}{\hbar}\,\mathrm{Im}\left[\left(\frac{-\imath}{\hbar}\right)^2\langle i|[[\mu(t),H(t)],H(t)]|q\rangle\right], \tag{15}$$

and similar expressions can be produced for the higher order derivatives. This hierarchy of expressions involves matrix elements $\langle i|\cdots|q\rangle$ of all combinations of commutators of μ and H_0. These commutators are the operators needed to establish controllability for the quantum system [31]. In particular, if the rank of the algebra formed by this hierarchy of operators has dimension N^2 (or N^2-1 for a traceless H_0), then the system is fully controllable. All that is required in the present case is that the arbitrary state $|q\rangle$ be fully reachable from state $|i\rangle$, which is demanded by the original assumption that the system is controllable. Reaching $|q\rangle$ will only happen if the commutators satisfy the rank condition, which implies that Eq. (11) cannot be satisfied for all time. Therefore, a contradiction exists and we may conclude that the variational condition in Eq. (6) cannot satisfy $\delta J/\delta\varepsilon(t)=0,\ \forall t$ for an arbitrary control field, which does not either satisfy $P_{i\to f}=0$ or $P_{i\to f}=1.0$. As a final comment, note that Eq. (11) will automatically be satisfied under the conditions in Sec. II A 1 as $|q\rangle=0$, and in Sec. II A 2 where $|q\rangle=|i\rangle$ (e.g., since $[\mu(t),H(t)]$ is an anti-Hermitian operator and $[[\mu(t),H(t)],H(t)]$ is a Hermitian operator, then the corresponding right-hand sides of Eqs. (14) and (15) have no imaginary parts, etc.)

This analysis may be alternatively argued by rewriting $\delta J/\delta\varepsilon(t)$ as

$$\begin{aligned}\frac{\delta J}{\delta\varepsilon(t)} &= \frac{2}{\hbar}\,\mathrm{Im}\left(\sum_{k=1}^{N}\langle i|\mu(t)|k\rangle\langle k|U^\dagger(T,0)|f\rangle\langle f|U(T,0)|i\rangle\right)\\ &= \frac{2}{\hbar}\,\mathrm{Im}[\langle i|\mu(t)|i\rangle\langle i|U^\dagger(T,0)|f\rangle\langle f|U(T,0)|i\rangle]\\ &\quad+\frac{2}{\hbar}\,\mathrm{Im}\left[\left(\sum_{k\neq i}\langle i|\mu(t)|k\rangle\langle k|U^\dagger(T,0)|f\rangle\right)\langle f|U(T,0)|i\rangle\right]\\ &= \frac{2}{\hbar}\,\mathrm{Im}\left[\left(\sum_{k\neq i}\langle i|\mu(t)|k\rangle\langle k|U^\dagger(T,0)|f\rangle\right)\langle f|U(T,0)|i\rangle\right]\end{aligned} \tag{16}$$

which is equal to zero for all t, if and only if either (i) the matrix element $\langle f|U(T,0)|i\rangle=0$ or (ii) the sum $\Sigma_{k\neq i}\langle i|\mu(t)|k\rangle\langle k|U^\dagger(T,0)|f\rangle=0$. The former case corresponds to $P_{i\to f}=0$, whereas the latter case leads immediately to the result that $\langle k|U^\dagger(T,0)|f\rangle=0,\ \forall k\neq i$, with the reasonable assumption that $\langle i|\mu(t)|k\rangle$ form a set of linearly independent functions of time for all k. Thus, in the second case we have that $P_{i\to f}=1.0-\Sigma_{k\neq i}|\langle k|U^\dagger(T,0)|f\rangle|^2=1.0$.

The conclusion we may draw from the collective analyses in Secs. II A 1, II A 2, and II A 3 is that the landscape in Eq. (2) only has extrema corresponding to null control $P_{i\to f}=0$ or perfect control $P_{i\to f}=1.0$. Particular control fields will be associated with each of these extrema values and no other control fields will satisfy the variational criterion $\delta J/\delta\varepsilon(t)=0,\ \forall t$ in Eq. (6). Thus, there are no false extrema in the control landscape at values $0<P_{i\to f}<1.0$, but the desirable extrema satisfying $P_{i\to f}=1.0$ will correspond to a null set of control fields $\varepsilon(t)$. This point is easily understood, and verified numerically, as randomly guessing a control field has no chance of producing the outcome $P_{i\to f}=1.0$. Nevertheless, almost any reasonable iterative procedure for solving Eqs. (4)–(6), especially a hill climbing technique, should be able to converge to $P_{i\to f}=1.0$ from an arbitrary initial trial field. This comment may at first appear to be only partially consistent with the many control simulations [16,20] where successful fields are readily found by a number of search algorithms, but rarely does one get close to a perfect yield (e.g., $P_{i\to f}>0.99$). The reason for this latter behavior is twofold. First, many of the computations in the literature include ancillary costs besides that in Eq. (3), and typically a fluence term $-w\int_0^T\varepsilon^2(t)dt$ is included with a weight $w>0$. In this case the variation in Eq. (6), becomes

$$\frac{\delta J}{\delta\varepsilon(t)} = \frac{2}{\hbar}\,\mathrm{Im}[\langle i|\mu(t)|q\rangle]-2w\varepsilon(t). \tag{17}$$

No longer can a perfect control solution be reached in accord with Eq. (9). This point follows from first assuming that perfect control is achieved that reduces Eq. (17) to $\delta J/\delta\varepsilon(t)=2w\varepsilon(t)$, which clearly cannot be equal to zero for all time. Even removing the fluence term and utilizing the original cost functional in Eq. (3) generally does not produce perfect control, $P_{i\to f}=1.0$, in numerical calculations for the additional reasons that (a) the target time T is typically fixed at a finite value, and (b) the control field $\varepsilon(t)$ is always constrained in one way or another at least minimally by some form of numerical discretization. However, careful numerical calculations [45] show that the final transition probabilities can readily be found satisfying the criteria of $P_{i\to f}>0.99$ by using the cost function in Eq. (3) with sufficiently large values for T (e.g., much larger than all natural time scales in the system) and discretizing the control $\varepsilon(t)$ with as much freedom as possible [e.g., creating a very fine mesh over time with the control variables being $\varepsilon_i=\varepsilon(t_i),\ i=1,2,\ldots$].

The above analysis was presented in the context of numerically performing optimal control designs, and the findings here clearly indicate that such future studies should continue to produce high quality yields (i.e., even perfect yields, when suitably performed, within machine precision and discretization errors). With this assurance of attaining quality control designs, the emphasis of such studies perhaps may be shifted more towards understanding mechanistic or other physical issues. But, ultimately the real purpose of quantum control is to achieve success in the laboratory, and adaptive learning control [8] is proving to be attractive for that purpose, even in systems of high complexity [22–29]. The findings in this section have a direct bearing on explaining the success of the growing numbers of these experiments, although the laboratory conditions are generally less well defined than in most of the numerical simulations of quantum control processes. The absolute final yields of the control experiments are typically not known, but the experiments collectively show that it is relatively easy to drive up the final yield to acceptable if not, dramatically enhanced values. This behavior occurs even in the common circumstances of the systems initially being in mixed states and exposed to some environmental decoherence during the control process. The control experiments also inherently involve some degree of laser, noise, and this issue will be returned to in Sec. II C. Finally, and perhaps most importantly, the experiments are carried out typically with highly constrained controls. This point is most evident from the fact that virtually all of the current experiments employ the Ti:sapphire laser system centered around 800 nm with initial unshaped pulses of ~25 nm bandwidth [29]. Such a "one control fits all dynamics" solution is surely a significant constraint. As with the numerical simulations, control field constraints inevitably reduce the iterative learning control process to taking a pathway across the control landscape that may be convoluted as well as quite likely not capable of achieving the desired goal (i.e., $P_{i\to f}=1.0$). Even modest constraints on the controls could yield what appears as artificial structure in the landscape (i.e., local suboptimal extrema), but which in fact corresponds to following a constrained path over the landscape. Importantly, the true landscape topology, as revealed in the analysis above, exists invariant to the constraints on the controls, just as controllability exists invariant to the same constraints being present. With these comments in mind, it is most encouraging that the present control experiments across broad classes of quantum phenomena appear so successful. The experimental outcomes should only get better as more flexible laser control capabilities (e.g., broader bandwidth sources) become available [46,47].

B. A bound on the landscape slope

The efficiency and stability of laboratory searches (or their computational counterparts) for effective quantum controls is a topic of prime concern. The magnitude of the landscape slope on the way towards an extremum is important in this regard. The following upper bound for $\delta J/\delta\varepsilon(t)$ can easily be established using Eq. (6),

$$\left\|\frac{\delta J}{\delta\varepsilon(t)}\right\| \le \frac{2}{\hbar}\|\mu\|, \tag{18}$$

where the unit norm of the time evolution operator was used. In any practical application, the norm of the dipole operator $\|\mu\|$ will always be finite. The bound in Eq. (18) has the simple physical interpretation that the landscape slope on the way towards an extremum is expected to be rather gentle without steep regions, thereby better assuring instability in the search effort. This behavior, along with the lack of false extrema, is attractive and suggests that the traditionally employed genetic algorithms for laboratory searching may be supplanted by other more efficient algorithms, possibly even including local ones, provided that due consideration is given to laboratory noise (e.g., sufficient signal averaging is employed).

C. Stability behavior in the neighborhood of an optimal control solution

The analysis in the preceding sections shows that upon searching over unconstrained control fields at least one may eventually be found such that $\delta J/\delta\varepsilon(t)=0$, $\forall t$, producing an ideal solution $P_{i\to f}=1.0$. A point of prime interest is the topology of the landscape in the vicinity of such an extremum, and this analysis is especially relevant for considerations of robustness to control field noise [48–50]. Robustness information may be extracted from the system's Hessian

$$\mathcal{H}(t,t') = \frac{\delta^2 J}{\delta\varepsilon(t')\delta\varepsilon(t)} \tag{19}$$

evaluated at the maximum of $P_{i\to f}=1.0$. The eigenvalues σ and eigenvectors $u(t')$ satisfying the integral equation

$$\int_0^T \mathcal{H}(t,t')u(t')dt' = \sigma u(t) \tag{20}$$

are of special interest. As the Hessian will be evaluated at a maximum, it follows that $\sigma\le 0$, and an analysis of the eigenvalues will be presented after first determining the Hessian.

Starting with Eq. (6), we may differentiate it once again to produce

$$\frac{\delta^2 J}{\delta\varepsilon(t')\delta\varepsilon(t)} = \mathrm{Im}\left(\left\langle \frac{\delta\lambda(t)}{\delta\varepsilon(t')}\right|\mu|\psi(t)\rangle + \langle\lambda(t)|\mu\left|\frac{\delta\psi(t)}{\delta\varepsilon(t')}\right\rangle\right), \quad t\ge t'. \tag{21}$$

The time restriction in Eq. (21) follows from causality conditions. The upper triangular portion of the Hessian in Eq. (21) is real, and the lower triangular portion is obtained by the transpose operation.

To evaluate Eq. (21), we first need $\delta|\psi(t)\rangle/\delta\varepsilon(t')$, which may be obtained by functionally differentiating Eq. (4) and noting that the time evolution operator $U(t,t')$ satisfies

$$\left(\imath\hbar\frac{\partial}{\partial t} - H_0 + \mu\varepsilon(t)\right)U(t,t') = 0, \quad U(t',t') = \mathbf{1} \tag{22}$$

to produce

$$\frac{\delta|\psi(t)\rangle}{\delta\varepsilon(t')} = \frac{\imath}{\hbar}U(t,t')\mu|\psi(t')\rangle. \tag{23}$$

Similarly, upon functionally differentiating Eq. (5) and solving the resulting differential equation we have

$$\frac{\delta|\lambda(t)\rangle}{\delta\varepsilon(t')} = -\frac{2\imath}{\hbar^2}U(t,T)|f\rangle\langle f|U(T,t')\mu\||\psi(t')\rangle, \quad t\ge t'. \tag{24}$$

As explained above, the analysis here will focus on the condition $t\ge t'$, and combining Eqs. (21), (23), and (24) produces the following result:

$$\frac{\delta^2 J}{\delta\varepsilon(t')\delta\varepsilon(t)} = -\frac{2}{\hbar^2}\,\mathrm{Re}[\langle i|U(0,T)|f\rangle \times\langle f|U(T,t)\mu U(t,t')\mu U(t',0)|i\rangle - \langle i|U(0,t')\mu U(t',T)|f\rangle\langle f|U(T,t)\mu U(t,0)|i\rangle]$$
$$= -\frac{2}{\hbar^2}\,\mathrm{Re}[\langle i|U(0,T)|f\rangle \times\langle f|U(T,t)\mu U(t,t')\mu U(t',0)|i\rangle - \langle i|U(0,t)\mu U(t,T)|f\rangle \times\langle f|U(T,t')\mu U(t',0)|i\rangle], \quad t\ge t', \tag{25}$$

by noting that $|\lambda(t)\rangle = -(2/\hbar)U(t,T)|f\rangle\langle f|U(T,0)|i\rangle$, from Eq. (5). Equations (19) and (25), and the symmetry relation

$$\mathcal{H}(t,t') = \mathcal{H}(t',t) \tag{26}$$

constitute the fundamental Hessian for analysis. To facilitate the discussions below, Eq. (25) can be further written as follows:

$$\mathcal{H}(t,t') = \frac{\delta^2 J}{\delta\varepsilon(t')\delta\varepsilon(t)}$$
$$= -\frac{2}{\hbar^2}\,\mathrm{Re}[\langle i|p\rangle\langle p|\mu(t)\mu(t')|i\rangle - \langle i|\mu(t)|p\rangle\langle p|\mu(t')|i\rangle]$$
$$= -\frac{2}{\hbar^2}\,\mathrm{Re}\sum_{k=1}^{N}\{[\langle i|p\rangle\langle p|\mu(t)|k\rangle - \langle i|\mu(t)|p\rangle\langle p|k\rangle]\langle k|\mu(t')|i\rangle\}$$
$$= -\frac{2}{\hbar^2}\,\mathrm{Re}\sum_{k\neq i}\{[\langle i|p\rangle\langle p|\mu(t)|k\rangle - \langle i|\mu(t)|p\rangle\langle p|k\rangle]\langle k|\mu(t')|i\rangle\}$$
$$= -\frac{2}{\hbar^2}\,\mathrm{Re}\sum_{k\neq i}\{\langle i|[|p\rangle\langle p|\mu(t) - \mu(t)|p\rangle\langle p|]|k\rangle\langle k|\mu(t')|i\rangle\}$$
$$= -\frac{2}{\hbar^2}\,\mathrm{Re}\sum_{k\neq i}\eta_k(t)\phi_k(t'), \quad t\ge t', \tag{27}$$

where we introduce $|p\rangle = U(0,T)|f\rangle$, $\eta_k(t) = |i\rangle(|p\rangle\langle p|\mu(t) - \mu(t)|p\rangle\langle p|)|k\rangle$, and $\phi_k(t) = \langle k|\mu(t)|i\rangle$. The Hessian expression in Eq. (27) is valid anywhere on the landscape. It is in general nondegenerate except for the two extreme cases, i.e., $P_{i\to f}=0$ and 1.0, considered in the following sections.

1. Hessian evaluation at a null solution: $P_{i\to f}=0$

In parallel with the analysis in Sec. II A 1 this case involves either $\varepsilon(t)=0$ or equivalently a nontrivial control field $\varepsilon(t)$ such that $\langle p|i\rangle = \langle f|U(T,0)|i\rangle = 0$ is satisfied. In either case, the first term in brackets in Eq. (25) is zero, which follows from the assumption $\langle f|i\rangle=0$. In the special case that $\varepsilon(t)=0$, corresponding to applying no control, Eq. (25) reduces to

$$\frac{\delta^2 J}{\delta\varepsilon(t')\delta\varepsilon(t)} = \frac{2}{\hbar^2}|\langle i|\mu|f\rangle|^2. \tag{28}$$

Similarly, the diagonal elements, $t=t'$, of the Hessian in Eq. (25) have the following form even for nontrivial control fields

$$\frac{\delta^2 J}{\delta[\varepsilon(t)]^2} = \frac{2}{\hbar^2}|\langle i|U(0,t)\mu U(t,T)|f\rangle|^2, \tag{29}$$

which is a positive quantity and reduces to Eq. (28) under the special case of $\varepsilon(t)=0$ and $t=t'$. From Eq. (29), it is evident that the trace of the Hessian $\int_0^T \mathcal{H}(t,t)dt$ is positive and bounded such that

$$\left|\int_0^T \mathcal{H}(t,t)dt\right| \le \frac{2T}{\hbar^2}\|\mu\|^2. \tag{30}$$

This relation implies the intuitive result that the rate of leaving the initial state is directly dictated by the norm of the dipole moment operator. Furthermore, it may be readily shown from Eq. (28) that the Hessian in the trivial case of $\varepsilon(t)=0$ has only one positive nonzero eigenvalue $\sigma = (2T/\hbar^2)|\langle i|\mu|f\rangle|^2$ with an eigenfunction, $u(t)=$constant. From Eq. (27), the case of a nontrivial null control, i.e., $\varepsilon(t)\neq 0$ for at least some times, produces

$$\mathcal{H}(t,t') = \frac{2}{\hbar^2}\,\mathrm{Re}[\langle i|\mu(t)|p\rangle\langle p|\mu(t')|i\rangle], \tag{31}$$

where $\langle p|i\rangle = \langle f|U(T,0)|i\rangle = 0$. The Hessian in Eq. (31) is a rank 2 operator with at most just two nonzero eigenvalues and eigenvectors. In this case, variations in two distinct directions around the minimum have particular effects associated with the nature of the original control field.

2. Stability analysis around a control maximum: $P_{i\to f}=1.0$

This case concerns evaluating the Hessian at a control that produces the desired maximum $P_{i\to f}=1.0$. The relation $|p\rangle\langle p| = U^\dagger(T,0)|f\rangle\langle f|U(T,0) = |i\rangle\langle i|$ may once again be employed to rewrite Eq. (25) in a physically transparent form using the dipole moment operator definition in Eq. (7),

$$\frac{\delta^2 J}{\delta\varepsilon(t')\delta\varepsilon(t)} = -\frac{2}{\hbar^2}\,\mathrm{Re}[\langle i|\mu(t)\mu(t')|i\rangle - \langle i|\mu(t)|i\rangle\langle i|\mu(t')|i\rangle]$$
$$= -\frac{2}{\hbar^2}\,\mathrm{Re}\langle i|\{[\mu(t) - \langle i|\mu(t)|i\rangle] \times[\mu(t') - \langle i|\mu(t')|i\rangle]\}|i\rangle, \quad t\ge t', \tag{32}$$

This result shows that the Hessian may be interpreted in terms of the covariance of the evolving dipole moment operators in the neighborhood of a transition probability maximum. Furthermore, the diagonal elements become

$$\frac{\delta^2 J}{\delta[\varepsilon(t)]^2} = -\frac{2}{\hbar^2}\langle i|[\mu(t) - \langle i|\mu(t)|i\rangle]^2|i\rangle$$

$$\geq -\frac{2}{\hbar^2}\langle i|\mu(t)\mu(t)|i\rangle$$

$$= -\frac{2}{\hbar^2}\langle \psi(t)|\mu^2|\psi(t)\rangle$$

$$\geq -\frac{2}{\hbar^2}\|\mu^2\|, \tag{33}$$

which is clearly negative, and the trace of the Hessian is bounded from below by the relation

$$\mathrm{Tr}\,\mathcal{H} = \int_0^T \mathcal{H}(t,t)dt \geq -\frac{2T}{\hbar^2}\|\mu^2\|, \tag{34}$$

noting that the norm $\|\mu^2\|$ should be finite in realistic applications. The remaining issue concerns the eigenvalue spectrum of the Hessian in Eq. (20).

A simple inspection of Eq. (27), recalling that $\langle p|k\rangle = \langle f|U(T,0)|k\rangle = 0\ \forall k \neq i$ and $|p\rangle\langle p| = |i\rangle\langle i|$ when $P_{i\to f}=1.0$ [i.e., producing $\eta_k(t) = \phi_k^*(t)$], shows that the Hessian in Eq. (20) at a transition probability maximum is a separable and symmetric kernel of the form

$$\mathcal{H}(t,t') = -\frac{2}{\hbar^2}\,\mathrm{Re}\sum_{k\neq i}\phi_k^*(t)\phi_k(t')$$

$$= -\frac{2}{\hbar^2}\sum_{k\neq i}[\phi_k^{\mathrm{Re}}(t)\times\phi_k^{\mathrm{Re}}(t') + \phi_k^{\mathrm{Im}}(t)\times\phi_k^{\mathrm{Im}}(t')], \tag{35}$$

where the $2N-2$ individual functions $\phi_k^{\mathrm{Re}}(t) = \mathrm{Re}[\langle k|\mu(t)|i\rangle]$ and $\phi_k^{\mathrm{Im}}(t) = \mathrm{Im}[\langle k|\mu(t)|i\rangle]$, $k=1,2,\ldots,i-1,i+1,\ldots,N$, come from the generally distinct matrix elements of the dipole operator $\mu(t)$. At most $2N-2$ of these functions can be linearly independent [51]. These functions may be reexpressed as a linearly independent orthogonal set, e.g., by suitable application of the Gram-Schmidt orthogonalization procedure [52]. From Eqs. (20) and (35), it is evident that

$$\int_0^T\int_0^T \mathcal{H}(t,t')u(t)u(t')dtdt'$$

$$= -\frac{2}{\hbar^2}\sum_{k\neq i}\left[\left(\int_0^T \phi_k^{\mathrm{Re}}(t)u(t)dt\right)^2 + \left(\int_0^T \phi_k^{\mathrm{Im}}(t)u(t)dt\right)^2\right]$$

$$\leq 0, \tag{36}$$

for any arbitrary continuous function $u(t)$, $t\in[0,T]$. Consequently, the finite rank Hessian $\mathcal{H}(t,t')$ at $P_{i\to f}=1.0$ is a continuous symmetric kernel function of negative semidefinite type, and it possesses at most $2N-2$ nonzero, negative eigenvalues $\sigma_1<0$, $\sigma_2<0,\ldots,\sigma_{2N-2}<0$ associated with $2N-2$ orthonormal eigenfunctions $u_1(t),u_2(t),\ldots,u_{2N-2}(t)$ [53]. In addition, the Hessian at $P_{i\to f}=1.0$ has infinitely many zero eigenvalues and associated eigenfunctions $u_0^i(t)$ satisfying $\langle\phi_k^{\mathrm{Re}}(t)|u_0^i(t)\rangle=0$ and $\langle\phi_k^{\mathrm{Im}}(t)|u_0^i(t)\rangle=0$. As the sum of these eigenvalues (i.e., the trace of the Hessian) is bounded by $-(2T/\hbar^2)\|\mu^2\|$ from Eq. (34), it is evident that as the Hilbert space dimension N increases, each individual nonzero eigenvalue will likely take on an ever smaller value, i.e., the average eigenvalue of the Hessian falls off as $\sim\frac{1}{2N-2}$. Furthermore, it is readily seen that around the control maximum $P_{i\to f}=1.0$, any small perturbation (i.e., noise) $\delta\varepsilon(t)$ in the control field $\varepsilon(t)$ yields a deviation $\delta P_{i\to f}$ in the optimal yield

$$\delta P_{i\to f} \approx \frac{1}{2}\int_0^T\int_0^T \delta\varepsilon(t)\mathcal{H}(t,t')\delta\varepsilon(t')dtdt'$$

$$= -\frac{1}{2}\int_0^T\int_0^T \delta\varepsilon(t)\left(\sum_{i=1}^{2N-2}|\sigma_i|u_i(t)u_i(t')\right)\delta\varepsilon(t')dtdt'$$

$$= -\frac{1}{2}\sum_{i=1}^{2N-2}|\sigma_i|(\delta\varepsilon,u_i)^2$$

$$\approx -\frac{1}{2}\frac{\sigma_{\delta\varepsilon}^2}{(2N-2)}\sum_{i=1}^{2N-2}|\sigma_i|$$

$$\geq -\frac{T\|\mu^2\|}{\hbar^2}\frac{\sigma_{\delta\varepsilon}^2}{(2N-2)}, \tag{37}$$

where $(\delta\varepsilon,u_i)=\int_0^T\delta\varepsilon(t)u_i(t)dt$ is the projection of the control field noise $\delta\varepsilon(t)$ in the direction of the ith eigenfunction $u_i(t)$ of the Hessian $\mathcal{H}(t,t')$ given in Eq. (35) and $\sigma_{\delta\varepsilon}^2=(\delta\varepsilon,\delta\varepsilon)=\Sigma_{i=1}^{2N-2}(\delta\varepsilon,u_i)^2$ is the mean square value of $\delta\varepsilon(t)$. The interpretation of the result in Eq. (37) calls for consideration of whether $\sigma_{\delta\varepsilon}^2$ likely has a dependence on N, especially as N rises. In the next to the last step in Eq. (37) the relation $(\delta\varepsilon,u_i)^2\approx\sigma_{\delta\varepsilon}^2/(2N-2)$ was utilized based on the reasonable assumption that the control field noise is expected to be equally dispersed along any of the eigenvectors of the Hessian [54]. However, in principle, $\sigma_{\delta\varepsilon}^2$ could depend on N, considering that more energy in the control could be required to manipulate systems of higher complexity (i.e., larger N). Practical considerations in realistic applications likely will call for a limited use of laser energy regardless of the system Hilbert space dimension N, which is consistent with the increasing success of controlling ever more complex systems [22–29]. Thus, it is reasonable to expect that $\sigma_{\delta\varepsilon}^2$ is essentially a system invariant constant or at most slowly varying in N. To this end, we may conclude the important result from Eq. (37) that control solutions at the maximum value of $P_{i\to f}=1.0$ have an inherent degree of robustness, which also tends to increase (or in the worst case remain neutrally stable) as the Hilbert space dimension rises. This behavior is very attractive for practical control, as noise is inevitably present in the laboratory.

III. CONCLUSION

This paper presents a quantum control landscape analysis directly in terms of the physically relevant control field including an elaboration of the topology around the null and maximal solutions to the variational equations. The basic conclusions of this work are coincident with the more abstract previous analysis utilizing the action matrix representation of the controlled dynamics [30]. Previous work also came to the natural conclusion that many, or even an infinite number of control maxima may be expected to exist [43,44]. This paper clearly shows that the landscape corresponding to these extrema has no false features. Furthermore, the slopes on the maxima and the curvature at the maxima solutions are both bound by the magnitude of the transition dipole moment. The trace of the Hessian being bounded from below has the important feature of implying robustness, and likely of an enhanced degree, with increasing Hilbert space dimension. In general, all of these results reveal the existence of remarkably attractive quantum control landscapes where the search efforts will encounter gentle slopes as well as maxima that are robustly flat. This behavior suggests that various, even simple, algorithms should be able to search through the accessible controls to find viable solutions, and second, a respectable degree of robustness to laboratory noise should exist [55]. The many successful control experiments are certainly consistent with this analysis.

The simple topology of the $P_{i\to f}$ landscape indicates that perhaps the main limiting factor in the control of quantum systems is the presence of constraints restricting access to the controls. That is, despite the fact that the landscape has an ideal topology, the presence of constraints could easily lead to tortuous search pathways across the landscape, which might artificially introduce multiple local extrema. These latter extrema would be reflective of the control constraints rather than the inherent structure of underlying the landscape. Other physical issues will certainly also enter, including the system being at finite temperature and exposed to decoherence during the dynamics. As with control noise, the presence of decoherence, at least of a weak nature, can be viewed as producing a lower resolution landscape rather than a fundamental change in its topological features [30]. However, starting out at a finite temperature, implies that the system contains entropy, which only could be removed by interaction with suitable bath. A landscape analysis based on starting with an arbitrary mixed initial state, as well as for the case of directly controlling unitary transformations themselves, has been carried out using the action matrix representation [56]. A similar analysis to that in this paper based on working with the control field $\varepsilon(t)$ should be insightful for these cases as well.

In summary, this work presents the basis to qualitatively understand the many successful optimal control simulations and the growing number of successful experiments. Perhaps most importantly, the conclusions from the generic topology of the quantum control landscapes provides the foundation to project ahead that many more positive quantum control experimental outcomes may be expected, even in manipulating complex systems. Having adequate controls is a central issue in executing the experiments to take advantage of the simple landscape topology. The many laboratory control successes, often with very constrained controls, bodes well for even better results in the future as the control sources improve.

ACKNOWLEDGMENTS

This work was partially supported by the Department of Energy and the Defense Advanced Research Projects Agency. The authors would like to thank Jason Dominy for his critical reading of the paper and valuable comments.

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