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# Characterization of the critical submanifolds in quantum ensemble control landscapes

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## Abstract

The quantum control landscape is defined as the functional that maps the control variables to the expectation values of an observable over the ensemble of quantum systems. Analyzing the topology of such landscapes is important for understanding the origins of the increasing number of laboratory successes in the optimal control of quantum processes. This paper proposes a simple scheme to compute the characteristics of the critical topology of the quantum ensemble control landscapes showing that the set of disjoint critical submanifolds one-to-one corresponds to a finite number of contingency tables that solely depend on the degeneracy structure of the eigenvalues of the initial system density matrix and the observable whose expectation value is to be maximized. The landscape characteristics can be calculated as functions of the table entries, including the dimensions and the numbers of positive and negative eigenvalues of the Hessian quadratic form of each of the connected components of the critical submanifolds. Typical examples are given to illustrate the effectiveness of this method.

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## 1. Introduction

The control of physical and chemical quantum-mechanical processes has [1, 2] recently seen many laboratory successes, especially utilizing ultrafast shaped laser pulses as controls. In many experiments, adaptive learning algorithms are applied to seek optimal control fields [3–5]. These achievements collectively reveal a surprising ease in discovering effective control fields despite the presence of noise, imperfections and severe constraints on the controls (i.e., limited bandwidth of  $\sim 20$  nm operating at  $\sim 800$  nm central wavelength in virtually all of the experiments). This behavior suggests that some generic foundation must lie behind the ease of teaching a laser to guide quantum system dynamics to produce specified outcomes.

Recently, the notion of quantum control landscapes was put forth to address this matter, where the landscape is the map from the space of control fields to some physical objective (e.g., quantum state transition probability [6, 7], the expectation value of a quantum observable [8, 9], or the fidelity of quantum gates [10]). The landscape critical topology (i.e., the topology of the set of critical points of the landscape) was analyzed to give insight into the effort to search for an optimal control. The initial exploration [6] of the landscape for the control of the state-to-state transition probability found that the landscape had only perfect extrema such that no false traps (i.e., local suboptima in addition to the global optimum) exist for the control searches to be caught in, which provides a basic understanding of the ease of searching for optimal controls.

The same feature exists for generalized landscapes of the observable expectation value on quantum ensembles [8, 9] except for up to  $\sim N!$  saddle suboptima, where  $N$  is the dimension of the system. The details of these critical points depend on the observable and the system initial state. For real quantum systems, especially molecular systems whose electronic, vibrational, and rotational states are involved, it is essential to investigate the effects of degeneracies upon the landscape topology to understand which classes of quantum control problems are expected to be successfully treated in the laboratory. Moreover, the landscape complexity needs to be considered when analyzing the scaling of control field search effort with  $N$ . This paper delves further into the control landscape topology for general finite-level quantum systems with arbitrary degeneracies in the statistical distribution of the initial state and the spectral structure of the observable.

The paper is organized as follows. Section 2 presents a group theoretical analysis of the quantum ensemble landscape in terms of double cosets, as a generalization of torii obtained in [8]. Section 3 ascribes the determination of double cosets to the enumeration of contingency tables whose marginal constraints are the degeneracy indices of the observable and the system initial density matrix. Section 4 uses these contingency tables to calculate several important topological features of the landscape critical submanifolds and discusses the physical implications of the findings. Section 5 provides some simple illustrative examples, and conclusions are presented in section 6.

## 2. The quantum ensemble control landscape

For an  $N$ -level quantum system with non-dissipative dynamics, the evolution of its density matrix is described by  $\rho(t) = U(t)\rho U^\dagger(t)$ , where  $\rho$  is the initial state and the unitary system propagator  $U(t)$  obeys the Schrödinger equation

$$i\hbar \frac{d}{dt} U(t) = H[\varepsilon(t)]U(t), \quad U(t_0) = I. \quad (1)$$

Here, the evolution extends over the interval  $0 \leq t \leq T$ . The governing Hamiltonian  $H$  contains a time-dependent control field  $\varepsilon(t)$  that guides the evolution of the quantum system. The goal is to find control fields that maximize the expectation value of some desired quantum observable  $\theta$  of the system at the time  $T$ :

$$J[\varepsilon(\cdot)] = \text{tr}\{U(T)\rho U^\dagger(T)\theta\}. \quad (2)$$

The optimization also can be defined for a non-Hermitian operator  $\theta$  [11], but here only the Hermitian cases will be studied. The quantum control landscape is defined as the mapping  $J: \varepsilon(\cdot) \rightarrow \mathbb{R}$  from the set of all admissible controls into the real values of  $J$ . Accordingly, the optimal controls must be critical points of  $J$  in  $\varepsilon(\cdot)$ -space. The main objective of the landscape analysis is to identify the geometry of all possible critical points, as they specify the accessible stationary values of  $J$  upon variation of the control. A special issue of concern is

the determination of whether false suboptimal critical point traps exist, as their presence could limit attempts to identify where to reach the absolute maximum of the landscape.

As the landscape over the space of control fields is complex to analyze, a natural approach is to re-express the analysis on the unitary group:

$$J(U) = \text{tr}(U\rho U^\dagger\theta), \quad U \in \mathcal{U}(N), \quad (3)$$

whose critical topology is easier to determine. The relationship between (2) and (3) is manifested by the chain rule

$$\delta J = \langle \nabla J(U(T)), \delta U(T) \rangle,$$

where  $\nabla J(U(T))$  is the gradient of  $J$  at  $U(T)$  in  $\mathcal{U}(N)$ , and  $\delta U(T)$  is the variation of  $U(T)$  caused by a control variation  $\delta\varepsilon(\cdot)$  over  $[0, T]$ . The bilinear operation  $\langle A, B \rangle = \text{Re tr}(A^\dagger B)$  is a Riemannian metric defined on  $\mathcal{U}(N)$ . A control  $\varepsilon(\cdot)$  is called regular if any variation  $\delta U(T)$  of the resulting  $U(T)$  is attainable by some admissible control variation, otherwise it is called singular [12]. For any regular control, the sufficient and necessary condition for  $\delta J \equiv 0$  upon any control variation is  $\nabla J(U(T)) = 0$ , which implies that it is critical if and only if the corresponding  $U(T)$  is critical for (3). Moreover, the optimality status (i.e., as a minimum, maximum or a saddle point) of the regular critical points and their corresponding  $U(T)$  are identical (see proof in [13]). Singular controls can also become critical points of (2) when the corresponding gradient vector  $\nabla J(U(T))$  is orthogonal to all attainable variations  $\delta U(T)$ , but in such cases  $\nabla J(U(T))$  is not necessarily vanishing. In this regard, the landscape critical points can be classified as regular and singular critical points, respectively. For landscape studies, it is more important to assess the universal properties that are Hamiltonian independent, which are essential for understanding the mounting successes in the control of various quantum systems. Hence, we are mainly concerned with the set of regular controls, for which the reduction from  $\varepsilon(\cdot)$  to  $U(T)$  preserves the critical topology from the  $\varepsilon(\cdot)$ -space to  $\mathcal{U}(N)$ . The influence of singular controls will be contained in another work.

The necessary and sufficient condition for  $U$  to be a critical point of (3) has been derived in [8, 14]. The basic concept is that, under the parametrization  $U(s, A) = e^{isA}U$ , where  $s \in \mathbb{R}$  and  $A^\dagger = A$ , of a neighborhood of  $U$  in  $\mathcal{U}(N)$ ,  $U$  is critical if and only if

$$\left. \frac{d}{ds} J[U(s, A)] \right|_{s=0} = 0, \quad \forall A^\dagger = A,$$

which gives

$$\text{tr}(iA[\theta, U\rho U^\dagger]) = 0, \quad \forall A^\dagger = A, \quad (4)$$

and this is equivalent to

$$[\theta, U\rho U^\dagger] = 0, \quad (5)$$

which can be used to describe the critical topology [6, 8, 11]. For systems with non-degenerate  $\rho$  and  $\theta$ , the critical submanifolds consist of  $N!$  number of  $N$ -torii embedded in the unitary group  $\mathcal{U}(N)$  [6]. The presence of degeneracies in  $\rho$  or  $\theta$  will merge these torii into a smaller number of larger critical submanifolds, and their explicit description will be resolved in the following sections.

### 3. Characterization of the critical submanifolds

Let  $R$  and  $S$  be the unitary transformations that diagonalize  $\rho$  and  $\theta$  respectively, i.e.,

$$\begin{aligned} \tilde{\rho} &= R^\dagger \rho R = \text{diag}\{\lambda_1, \dots, \lambda_1; \dots; \lambda_r, \dots, \lambda_r\}, \\ \tilde{\theta} &= S^\dagger \theta S = \text{diag}\{\epsilon_1, \dots, \epsilon_1; \dots; \epsilon_s, \dots, \epsilon_s\}, \end{aligned}$$

where  $\lambda_1 > \dots > \lambda_r$  are distinct eigenvalues of  $\rho$  with  $n_1, \dots, n_r$  multiplicities and  $\epsilon_1 > \dots > \epsilon_s$  are distinct eigenvalues of  $\theta$  with  $m_1, \dots, m_s$  multiplicities. The landscape functional can be written as

$$J(\tilde{U}) = \text{tr}(UR\tilde{\rho}R^\dagger U^\dagger S\tilde{\theta}S^\dagger) = \text{tr}[(S^\dagger UR)\tilde{\rho}(S^\dagger UR)^\dagger \tilde{\theta}] = \text{tr}(\tilde{U}\tilde{\rho}\tilde{U}^\dagger \tilde{\theta}), \quad (6)$$

where the automorphism  $\tilde{U} = S^\dagger UR$  also runs over  $\mathcal{U}(N)$ . Thus, without loss of generality, we can always assume that both  $\rho$  and  $\theta$  are diagonal.

**Theorem 1.** *Let  $\mathcal{U}(\mathbf{n})$  be the product group  $\mathcal{U}(n_1) \times \dots \times \mathcal{U}(n_r)$  where  $\mathcal{U}(n_i)$  is the  $n_i$ -dimensional unitary group acting on the eigenspace of  $\lambda_i$ , and  $\mathcal{U}(\mathbf{m}) = \mathcal{U}(m_1) \times \dots \times \mathcal{U}(m_s)$  is defined in the same manner relevant to  $\theta$ . A unitary matrix  $U$  is a critical point of (6) if and only if it is in the double coset*

$$\mathcal{U}(\mathbf{n})\pi\mathcal{U}(\mathbf{m}) = \{P\pi Q : P \in \mathcal{U}(\mathbf{m}), Q \in \mathcal{U}(\mathbf{n})\}$$

of some permutation matrix  $\pi$ .

**Proof.** Suppose that  $U$  is a critical point. According to (5),  $U$  must transform  $\rho$  into block-diagonal form, whose block lengths correspond to the degenerate subspace dimensions of  $\theta$ . This block-diagonal matrix  $U\rho U^\dagger$  can be subsequently diagonalized by a  $m_1 \times \dots \times m_s$  block-diagonal unitary matrix  $P \in \mathcal{U}(\mathbf{m})$ . Since unitary transformations will not alter the spectrum of  $\rho$ , the resulting diagonal matrix can always be written as  $\pi^\dagger \rho \pi$ , where  $\pi$  is a permutation matrix that reorders the diagonal elements of  $\rho$ . These operations can be expressed as  $P^\dagger U \rho U^\dagger P = \pi \rho \pi^\dagger$ , which is equivalent to

$$\rho = (\pi^\dagger P^\dagger U)\rho(\pi^\dagger P^\dagger U)^\dagger.$$

By definition, the matrix  $Q = \pi^\dagger P^\dagger U$  must be an element of the stabilizer of  $\rho$  in  $\mathcal{U}(N)$ , which from group theory is identified with  $\mathcal{U}(\mathbf{n})$ . Thus, we obtain the decomposition  $U = P\pi Q$ . Conversely, one can easily verify that an arbitrary permutation matrix is critical,  $J(P\pi Q) = J(\pi)$  and the critical condition (5) is satisfied for both  $\pi$  and  $P\pi Q$ , implying that  $P\pi Q$  is also critical producing the same landscape value as  $\pi$ .  $\square$

Theorem 1 shows that the critical manifold is a union of double cosets,

$$\mathcal{M} = \bigcup_{\pi \in \mathcal{P}(N)} \mathcal{U}(\mathbf{m})\pi\mathcal{U}(\mathbf{n}), \quad (7)$$

where  $\mathcal{P}(N)$  denotes the permutation group over  $N$  indices. The set  $\mathcal{M}_\pi = \mathcal{U}(\mathbf{m})\pi\mathcal{U}(\mathbf{n})$  is the double coset of  $\pi$  in  $\mathcal{U}(N)$  with respect to  $\mathcal{U}(\mathbf{m})$  and  $\mathcal{U}(\mathbf{n})$ . In the special case that both  $\rho$  and  $\theta$  are non-degenerate [8], the stabilizer of  $\rho$  and  $\theta$  are both products of  $N$  one-dimensional unitary groups, i.e.,

$$\mathcal{U}(\mathbf{m}) = \mathcal{U}(\mathbf{n}) = [\mathcal{U}(1)]^N,$$

the critical manifold as the double cosets can be verified to consist of  $N!$  disjoint  $N$ -torii, each of which is labeled by a permutation matrix. Generally, the occurrence of degeneracies in  $\rho$  and  $\theta$  will ‘merge’ two torii  $\mathcal{M}_\pi$  and  $\mathcal{M}_{\pi'}$  together if  $\pi'$  happens to be in  $\mathcal{M}_\pi$ , thereby reducing the number of original disjoint critical submanifolds, but increasing their dimensions. These critical submanifolds can be taken as the generalized ‘torii’. Let  $\tilde{\mathcal{P}}$  be the set of all inequivalent partitions of  $\mathcal{P}(N)$  with respect to the equivalence relation ‘ $\sim$ ’ on  $\mathcal{P}(N)$ , which is defined as  $\pi' \sim \pi$  if  $\pi' \in \mathcal{M}_\pi$ . The characterization of the disjoint critical submanifolds can be identified as seeking all inequivalent double cosets of the permutation matrices in  $\mathcal{U}(N)$  with respect to  $\mathcal{U}(\mathbf{n})$  and  $\mathcal{U}(\mathbf{m})$

$$\mathcal{M} = \bigcup_{\tilde{\pi} \in \tilde{\mathcal{P}}} \mathcal{U}(\mathbf{m})\tilde{\pi}\mathcal{U}(\mathbf{n}). \quad (8)$$

**Table 1.** Contingency table  $\mathbf{K}$ .

	$m_1$	$m_2$	$\cdots$	$m_s$
$n_1$	$k_{11}$	$k_{12}$	$\cdots$	$k_{1s}$
$n_2$	$k_{21}$	$k_{22}$	$\cdots$	$k_{2s}$
$\vdots$	$\vdots$	$\vdots$	$\ddots$	$\vdots$
$n_r$	$k_{r1}$	$k_{r2}$	$\cdots$	$k_{rs}$

**Table 2.** The contingency table for the illustrative three-level system.

	2	1
1	1	0
2	1	1

We now introduce the concept of a contingency table to simplify the abstract description of a critical submanifold expressed as the double coset of some permutation  $\pi$ . Suppose that both  $\rho$  and  $\theta$  have their diagonal elements arranged in decreasing order. Let  $k_{ij}$  be the number of positions on the diagonal, where the eigenvalues  $\lambda_i$  and  $\epsilon_j$  appear simultaneously after imposing the permutation  $\pi$  on  $\theta$ . The contingency table consists of these non-negative *overlap numbers* as arranged in table 1, whose row sums are  $n_i$ ’s and column sums are  $m_j$ ’s.

For example, for a three-level quantum system where  $\rho = \text{diag}\{0.4, 0.3, 0.3\}$  and  $\theta = \text{diag}\{0.4, 0.4, 0.2\}$ , the corresponding marginal constraints are  $n_1 = 1$  and  $n_2 = 2$  for row sums;  $m_1 = 2$  and  $m_2 = 1$  for column sums. Suppose that a permutation transformation  $\pi$  exchanges the second and third eigenvalues of  $\theta$ , i.e.,  $\theta' = \pi\theta\pi^\dagger = \text{diag}\{0.4, 0.2, 0.4\}$ . Then the overlap number of the first eigenvalue 0.4 of  $\rho$  with the first eigenvalue 0.4 of  $\theta$  is  $k_{11} = 1$ , which appears at the first position on the diagonal of  $\rho$  and  $\theta'$ ;  $k_{22} = 1$  is the overlap number of the second eigenvalue 0.3 of  $\rho$  with the second eigenvalue 0.2 of  $\theta$ , which can be seen from the second diagonal elements of  $\rho$  and  $\theta'$ . Similarly,  $k_{21} = 1$  and  $k_{12} = 0$ . The resulting contingency table is shown in table 2.

Every permutation matrix leads to a contingency table, but the same contingency table may be produced from different permutation matrices which are mutually equivalent with respect to the relation ‘ $\sim$ ’. Therefore, the contingency tables provide an equivalent description of the critical submanifolds.

**Theorem 2.** Every critical submanifold of (3) is uniquely determined by a contingency table satisfying the above marginal conditions, and vice versa. The critical submanifold corresponding to a contingency table  $\mathbf{K}$  can be expressed as the quotient set

$$\mathcal{M}_{\mathbf{K}} = \frac{\mathcal{U}(\mathbf{m}) \times \mathcal{U}(\mathbf{n})}{\mathcal{U}(\mathbf{K})}, \quad \text{where} \quad \mathcal{U}(\mathbf{K}) = \prod_{k_{ij} \neq 0} \mathcal{U}(k_{ij}). \quad (9)$$

**Proof.** Define  $F_\pi(P, Q) = P\pi Q$ , where  $(P, Q) \in \mathcal{U}(\mathbf{m}) \times \mathcal{U}(\mathbf{n})$ , as the bilateral operation on  $\pi$ . The set of critical points that are equivalent with  $\pi$  is characterized by the stabilizer  $\text{stab}(\pi)$  of  $F_\pi$  in  $\mathcal{U}(\mathbf{m}) \times \mathcal{U}(\mathbf{n})$ , i.e., the set of matrix pairs  $(U, V) \in \mathcal{U}(\mathbf{m}) \times \mathcal{U}(\mathbf{n})$  such that  $F_\pi(U, V) = U\pi V = \pi$ . Hence, the critical submanifold  $\mathcal{M}_\pi$  can be identified as the quotient set of  $\mathcal{U}(\mathbf{m}) \times \mathcal{U}(\mathbf{n})$  divided by  $\text{stab}(\pi)$ , which is isomorphic to the set  $\mathcal{U}(\mathbf{n}) \cap \pi^\dagger \mathcal{U}(\mathbf{m}) \pi$  as follows:

$$\text{stab}(\pi) = \{(\pi V^\dagger \pi^\dagger, V) : V \in \mathcal{U}(\mathbf{n}) \cap \pi^\dagger \mathcal{U}(\mathbf{m}) \pi\}.$$

The intersection set  $\mathcal{U}(\mathbf{n}) \cap \pi^\dagger \mathcal{U}(\mathbf{m}) \pi$  is a Lie subgroup of  $\mathcal{U}(N)$  and can be decomposed into the product of smaller unitary groups as  $\mathcal{U}(\mathbf{K}) = \mathcal{U}(k_{11}) \times \cdots \times \mathcal{U}(k_{rs})$ , where the  $k_{ij}$  s are the entries of the contingency table for  $\pi$ . This leads to the expression (9). Therefore, every critical submanifold can be uniquely determined by a contingency table. On the other hand, given the contingency table shown in table 1, one can shuffle the originally ordered diagonal elements of  $\theta$  such that  $k_{ij}$  of the  $m_j$  eigenvalues  $\epsilon_j$  are moved to any of the  $n_i$  positions where the eigenvalues  $\lambda_i$  of  $\rho$  are located. Any of such shuffles can be represented by a permutation operation corresponding to this table. Hence, any contingency table corresponds to at least one critical submanifold. In this manner, we prove the one-to-one correspondence between critical submanifolds and contingency tables.  $\square$

The analysis above provides an easy means to analyze the critical topology of general quantum ensemble landscapes, by which the evaluation of the critical submanifolds on continuous Lie groups is reduced to a simple finite combinatorial problem solvable by a computer. From now on, we use the contingency tables to label the different branches of the critical submanifolds. The landscape value of the critical submanifold  $\mathcal{M}_{\mathbf{K}}$  corresponding to contingency table  $\mathbf{K}$  is

$$J(\mathbf{K}) = \sum_{i=1}^N \rho_{ii} \theta_{\pi(i)\pi(i)} = \sum_{i=1}^r \sum_{j=1}^s k_{ij} \lambda_i \epsilon_j,$$

where  $\pi$  is some permutation matrix whose contingency table is  $\mathbf{K}$ .

#### 4. Characteristics of the critical submanifolds

The contingency tables are not only convenient for labeling the critical submanifolds but also powerful in identifying their intrinsic topological characteristics that are important for the performance of search algorithms seeking effective quantum optimal control field. These characteristics include: (1) the dimension,  $D_0(\mathbf{K})$ , for a given critical submanifold  $\mathcal{M}_{\mathbf{K}}$ , which qualitatively reflects the size of the associated critical submanifold (especially the maximum of the landscape) that is crucial for assessing robustness to control field noise; (2) the numbers of the positive and negative ‘principal axis directions’ near a critical submanifold,  $D_+(\mathbf{K})$  and  $D_-(\mathbf{K})$  [6, 8], which can affect the path taken by the search algorithm through the influence of the number of positive and negative directions in the vicinity of suboptimal regions.

The dimension  $D_0(\mathbf{K})$  in terms of the associated contingency table  $\mathbf{K}$  can be calculated in an easier way by (9), with the fact that the dimension of  $U(\ell)$  is  $\ell^2$ :

$$\begin{aligned} D_0(\mathbf{K}) &= \dim[\mathcal{U}(\mathbf{n})] + \dim[\mathcal{U}(\mathbf{m})] - \dim[\mathcal{U}(\mathbf{K})] \\ &= \sum_{i=1}^r n_i^2 + \sum_{j=1}^s m_j^2 - \sum_{i=1}^r \sum_{j=1}^s k_{ij}^2. \end{aligned} \quad (10)$$

Computationally, the characteristics  $D_0(\mathbf{K})$  (resp.,  $D_+(\mathbf{K})$  and  $D_-(\mathbf{K})$ ) can be identified as the numbers of zero (resp., positive and negative) eigenvalues of the Hessian quadratic form (HQF) at any  $U \in \mathcal{M}_{\mathbf{K}}$ , which determines the geometry in the vicinity of  $U$ . The HQF is defined as the second-order term of  $A$  in the Taylor expansion of  $J(e^{iA}U)$  at  $U$ , and it can be easily obtained as follows:

$$\mathcal{H}(A) = \text{tr}(AU\rho U^\dagger A\theta - A^2U\rho U^\dagger \theta).$$

Let  $x_{\beta\gamma}$  and  $y_{\beta\gamma}$  be the real and imaginary parts of the  $\beta\gamma$ th matrix elements of  $A$ , which represent the coordinate variables in the tangent space of  $U$ . Then the Hessian form can be further expanded as the following sum [8]:

$$\mathcal{H}(A) = - \sum_{1 \leq \beta < \gamma \leq N} (\lambda_\beta - \lambda_\gamma)(\epsilon_\beta - \epsilon_\gamma)(x_{\beta\gamma}^2 + y_{\beta\gamma}^2), \quad (11)$$

where the real numbers  $\lambda_\beta$  and  $\lambda_\gamma$  ( $\beta, \gamma = 1, \dots, N$ ) are the rearranged  $N$  diagonal elements of  $\rho$  and  $\theta$  after some permutation operation whose contingency table is  $\mathbf{K}$ . Since there are always  $N^2$  terms in (11), the sum of the three indices should satisfy

$$D_0(\mathbf{K}) + D_+(\mathbf{K}) + D_-(\mathbf{K}) = N^2.$$

According to (11), the index  $D_+$  is twice the number of  $(\beta, \gamma)$  pairs for which  $(\lambda_\beta - \lambda_\gamma)(\epsilon_\beta - \epsilon_\gamma) < 0$ , while  $D_-$  is twice the number of  $(\beta, \gamma)$  pairs for which  $(\lambda_\beta - \lambda_\gamma)(\epsilon_\beta - \epsilon_\gamma) > 0$ . A critical submanifold is locally maximal (or minimal) if the corresponding index  $D_+ = 0$  (or  $D_- = 0$ ), i.e., the Hessian eigenvalues are all negative (or positive). This happens only if the magnitudes of the  $\lambda_\beta$ 's and the  $\epsilon_\gamma$ 's are in the same (or opposite) order, which corresponds to a unique maximal (or minimal) critical submanifold. Therefore, there exist no local suboptima over the control landscape. Besides the absolute maximal and minimal critical submanifolds, all other critical points are saddles. In practice, the saddles will never form false traps for a search algorithm to approach global optimal controls although the search may be slowed down when the algorithm runs in their neighborhoods. This behavior is a strong support for the observed relative ease of searching for optimal controls in quantum systems.

Now suppose that  $\lambda_\beta = \lambda_i$  and  $\lambda_\gamma = \lambda_p$  ( $1 \leq i, p \leq r$ );  $\epsilon_\beta = \epsilon_j$  and  $\epsilon_\gamma = \epsilon_q$  ( $1 \leq j, q \leq s$ ). By the nature of the contingency table, there are  $k_{ij}k_{pq}$  possibilities for this to happen. Since  $\lambda_1, \dots, \lambda_r$  and  $\epsilon_1, \dots, \epsilon_s$  are both in decreasing order, we have the relationship:

$$(\lambda_\beta - \lambda_\gamma)(\epsilon_\beta - \epsilon_\gamma) \geq 0 \iff (i - p)(j - q) \geq 0.$$

Hence, the  $D_\pm(\mathbf{K})$  indices are equal to the following quadratic function of  $\mathbf{K}$

$$D_\pm(\mathbf{K}) = 2 \sum_{(i-p)(j-q) \leq 0} k_{ij}k_{pq} \quad (12)$$

which, by taking  $\mathbf{K}$  as a  $r \times s$  integer matrix, can be further written in a compact form:

$$D_+(\mathbf{K}) = 2 \text{tr}(\mathbf{J}_r \mathbf{K} \mathbf{J}_s \mathbf{K}^\dagger), \quad (13)$$

$$D_-(\mathbf{K}) = 2 \text{tr}(\mathbf{J}_r \mathbf{K} \mathbf{J}_s^\dagger \mathbf{K}^\dagger). \quad (14)$$

Here, the entries of the index matrix  $\mathbf{J}_t = \{\sigma_{ij}\}$  ( $1 \leq i, j \leq t$ ) are in the upper triangular form

$$\sigma_{ij} = \begin{cases} 1, & i < j, \\ 0, & i \geq j. \end{cases}$$

We have not found an explicit formula for counting the number of critical submanifolds, which mainly counts the saddle critical submanifolds because there is always only one maximal and minimal critical submanifold. Generally, the breaking of the degeneracies in  $\rho$  and  $\theta$  (e.g., when  $\rho$  turns from a pure state to a mixed state) increases the number of saddle critical submanifolds, which may have adverse impacts on the search algorithms by creating a tortured path to approach the global optima. The examples in section 5 analyze some special cases where the number of critical submanifolds can be explicitly calculated. For more complex situations, some good estimates have been found in [15, 16].

In real physical systems, there always exist disturbances or some other factors that may destroy the degenerate structures of the initial density matrix and observable operator. Since all the topological landscape properties are uniquely determined by their degeneracy degrees, even a very small perturbation may alter the critical topology. Heuristically, supposing the eigenvalues of  $\rho$  and  $\theta$  are well separated, the perturbation will wrinkle the existing ‘flat’ critical submanifolds such that new smaller ones emerge, increasing the number of critical submanifolds up to  $N!$ , while retaining only two absolute extrema. The submanifold dimensions and principal axis directions are consequently different in these cases. Nevertheless, provided that the perturbation is sufficiently small, e.g., compared with the step size taken upon numerical or experimental searching for a control, we expect that the structural changes of the critical topology will have little influence on the optimization algorithm performance because the original critical regions will remain almost flat. To obtain a full view of the landscape, the geometrical curvature that reflects ‘steepness’ (or ‘flatness’) of the landscape near the critical submanifolds should also be considered.

## 5. Examples

In this section we will apply the results obtained above to several simple examples. to illustrate some basic features of the control landscape topology.

### 5.1. The pure state system

Suppose that  $\rho$  is a pure state, then  $n_1 = 1$  for  $\lambda_1 = 1$  and  $n_2 = N - 1$  for  $\lambda_2 = 0$ ; the observable  $\theta$  has  $r$  distinct eigenvalues  $\epsilon_1 > \dots > \epsilon_s$  with degeneracies being  $m_1, \dots, m_s$ . The enumeration of the contingency tables is rather simple because the positions in only the first row can be filled with a single 1 and  $s - 1$  zeros, which amounts to  $s$  different contingency tables as shown in table 2. Therefore, there are  $s$  distinct critical submanifolds for such pure-state systems. Applying (10) and (12), the dimension and  $D_{\pm}(\mathbf{K}_j)$  indices of the  $j$ th critical submanifold are

$$\begin{aligned} D_0(\mathbf{K}_j) &= N(N - 2) + 2m_j, \\ D_+(\mathbf{K}_j) &= 2(m_1 + \dots + m_{j-1}), \\ D_-(\mathbf{K}_j) &= 2(m_{j+1} + \dots + m_s), \end{aligned}$$

for  $j = 1, \dots, s$ . One can specify that  $\mathcal{M}_{\mathbf{K}_1}$  is the maximum submanifold with

$$D_0(\mathbf{K}_1) = N(N - 2) + 2m_1, \quad D_-(\mathbf{K}_1) = 2(N - m_1);$$

$\mathcal{M}_{\mathbf{K}_s}$  is the minimum submanifold with

$$D_0(\mathbf{K}_s) = N(N - 2) + 2m_s, \quad D_+(\mathbf{K}_s) = 2(N - m_s).$$

Obviously, high degeneracy in the largest eigenvalue of the observable may facilitate the optimal searches toward maximizing the cost functional and enhance the robustness of perfect control, while that of the smaller eigenvalues retards the control searching on level sets of lower landscape values.

### 5.2. Non-degenerate observable

Suppose the spectrum of  $\theta$  is fully non-degenerate, i.e.,  $m_1 = \dots = m_N = 1$ ; the density matrix  $\rho$  has  $r$  distinct eigenvalues  $\lambda_1 > \dots > \lambda_r$  with degeneracies being  $n_1, \dots, n_r$ . Then, the entries in the contingency table can only be 0 or 1, and there are  $\frac{N!}{n_1!(N-n_1)!}$  choices for

**Table 3.** Contingency tables  $\mathbf{K}_j$  for a pure state system,  $j = 1, \dots, s$ .

	$m_1$	$\dots$	$m_j$	$\dots$	$m_s$
1	0	$\dots$	1	$\dots$	0
$N - 1$	$m_1$	$\dots$	$m_j - 1$	$\dots$	$m_s$

placing  $n_1$  numbers 1 in the  $N$  positions in the first row. Having fixed the first row, there are  $\frac{(N-n_1)!}{n_2!(N-n_1-n_2)!}$  choices to place  $n_2$  numbers 1 in the remaining  $N - n_1$  positions in the second row, etc. Finally, the total number of distinct critical submanifolds is

$$\mathcal{N} = \frac{N!}{n_1!(N-n_1)!} \cdot \frac{(N-n_1)!}{n_2!(N-n_1-n_2)!} \cdots \frac{(N-n_1-\dots-n_{r-2})!}{n_{r-1}!n_r!} = \frac{N!}{n_1! \cdots n_r!}.$$

The corresponding dimension of each critical submanifold is

$$D_0(\mathbf{K}_\alpha) = \sum_{i=1}^r n_i^2, \quad \alpha = 1, \dots, \mathcal{N}.$$

From (9), these critical submanifolds are diffeomorphic to each other. This result generalizes the special case of both  $\rho$  and  $\theta$  being non-degenerate, whose critical submanifolds are all  $N$ -torii. The  $D_\pm(\mathbf{K})$  indices vary with the corresponding contingency tables, among which  $D_- = N^2 - \sum_i n_i^2$  at the maximum and  $D_+ = N^2 - \sum_i n_i^2$  at the minimum.

### 5.3. A multi-degenerate system

Consider an eight-level system with degeneracies  $\mathbf{n} = (1, 3, 4)$  and  $\mathbf{m} = (2, 6)$ . All of the possible contingency tables are listed as below

$$\mathbf{K}_1 = \begin{bmatrix} 0 & 1 \\ 0 & 3 \\ 2 & 2 \end{bmatrix}, \quad \mathbf{K}_2 = \begin{bmatrix} 0 & 1 \\ 1 & 2 \\ 1 & 3 \end{bmatrix}, \quad \mathbf{K}_3 = \begin{bmatrix} 1 & 0 \\ 0 & 3 \\ 1 & 3 \end{bmatrix},$$

$$\mathbf{K}_4 = \begin{bmatrix} 0 & 1 \\ 2 & 1 \\ 0 & 4 \end{bmatrix}, \quad \mathbf{K}_5 = \begin{bmatrix} 1 & 0 \\ 1 & 2 \\ 0 & 4 \end{bmatrix}.$$

The topological characteristics of the five critical submanifolds are summarized in table 4. The second critical submanifold is the largest with dimension 50. The minimum submanifold is the second largest. The maximum submanifold is the smallest, and from (9), it is diffeomorphic to the fourth saddle submanifold because they have the same group of nonzero entries in their contingency tables. The high degree of degeneracies yields the indicated high dimensions of the critical submanifolds.

### 5.4. Molecular systems

A typical molecular system at room temperature and under strong field bound-state control usually possesses a very large number  $N$  of accessible discrete levels. Suppose the system has initially populated several non-degenerate vibrational levels  $|v_1\rangle, \dots, |v_r\rangle$ , each of which has associated rotational states. Often the rotational states are densely packed, and for illustration here, they will be taken as degenerate with  $n_i$  ( $i = 1, \dots, r$ ) associated with each vibrational state  $|v_i\rangle$ . The population  $p_i$  of  $|v_i\rangle$  is then equally distributed over the  $n_i$  rotational states. As

**Table 4.** Characteristics for the case of an eight-level system with degeneracies  $\mathbf{n} = (1, 3, 4)$  and  $\mathbf{m} = (2, 6)$ .

No.	1	2	3	4	5
Manifold dimension	48	50	46	44	44
Positive axis direction	16	8	6	4	0
Negative axis direction	0	6	12	16	20
Type	Minimum	Saddle	Saddle	Saddle	Maximum

**Table 5.** Contingency tables for the molecular illustration.

	$n_1$	$\cdots$	$n_r$	$N_0$
$M$	$k_{11}$	$\cdots$	$k_{1r}$	$k_{1,r+1}$
$N - M$	$k_{21}$	$\cdots$	$k_{2r}$	$k_{2,r+1}$

the control field can import considerable energy into the molecule,  $N$  can be very large such that  $\sum_i n_i \ll N$ .

Consider the transition control from the initial state to an  $M$ -dimension subspace, where both  $M$  and  $N - M$  are larger than  $\sum_i n_i$ . The target observable is expressed as a projector

$$\theta = \frac{1}{M} \sum_{i=1}^M |q_i\rangle\langle q_i|,$$

on this  $M$ -dimensional subspace. The contingency tables corresponding to the critical submanifolds have to satisfy  $(r + 1)$  column-sum conditions  $n_1, \dots, n_r, N_0$ , where  $N_0 = N - \sum_{i=1}^r n_i$ , and two row-sum conditions  $M$  and  $N - M$ , as shown in table 5.

Since the contingency table is determined by the values of independent variables  $k_{11}, \dots, k_{1r}$  which can vary from 0 to  $n_i$  without violating the marginal condition, we can obtain the following number of critical submanifolds

$$\mathcal{N} = \prod_{i=1}^r (n_i + 1).$$

The global maximal submanifold corresponds to  $k_{1i} = n_i, i = 1, \dots, r$ , which from (10) has dimension

$$D_0 = N^2 - 2(N - M)(n_1 + \cdots + n_r).$$

Another special case is control to a specific final degenerate state. For simplicity, we assume that the degeneracies of the initially populated vibrational states are identical, i.e.,  $n_1 = \cdots = n_r = n$ . The projector  $\theta$  then acts on a  $n$ -dimensional subspace, i.e.,  $M = n$ . In this case, the number of critical submanifolds equals the number of nonnegative unordered partitions of  $n$  in the first row of table 4, which gives

$$\mathcal{N} = \frac{(n + r)!}{n!r!}.$$

In this case, the dimension of the global maximal manifold is  $D_0 = (N - n)^2 + n^2$ .

The molecular illustration above has (i) a small number of initially populated states, (ii) a target population with a modest number of states, and (iii) a very large number  $\mathcal{N}$  of accessible states. These circumstances can arise quite commonly in molecular control, and they produce the outcome that the global maximum submanifold dimension  $D_0$  is very large and scales

as  $\sim N^2$ . This behavior implies that finding a control on the maximum critical submanifold should be relatively easy due to the large size of the target submanifold and the lack of false traps.

## 6. Discussion

We have given a complete description of the critical topology of quantum ensemble landscapes for general finite-level systems over their propagator spaces. It is shown that the most important topological features can be calculated from a set of contingency tables that uniquely specify the critical submanifolds. This framework opens up the possibility of exploring the landscape of more complex quantum systems. We have applied it to the control landscape of open quantum systems [13], and another potential application in the future is the analyses of control landscapes for an infinite-dimensional quantum system as the limit of a series of finite-dimensional systems, which can be dealt with using the techniques developed here.

The landscape topology is based on a strong assumption that the system is fully controllable, which may be limited in practice. In particular, a broad class of partially controllable systems have their propagators constrained on a proper subgroup  $\mathcal{G}$  of  $\mathcal{U}(N)$ , and the condition for some  $U \in \mathcal{G}$  to be critical is reduced to

$$\text{tr}(A[\theta, U\rho U^\dagger]) = 0, \quad \forall A \in \mathfrak{g}, \quad (15)$$

where  $\mathfrak{g}$  is the Lie algebra of  $\mathcal{G}$ . Obviously, every critical point of the landscape on  $\mathcal{U}(N)$  that belongs to  $\mathcal{G}$  still remains critical on  $\mathcal{G}$  (there may exist additional critical points since the condition (15) is weaker than (4)). In certain cases, these critical points may degenerate from the original saddle points into local maximal (minimal) points. Hence, the results obtained here for fully controllable systems provide a basis for future landscapes studies of uncontrollable systems.

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## References

- [1] Brixner T, Damrauer N H, Krampert G, Niklaus P and Gerber G 2003 Femtosecond learning control of quantum dynamics in gases and liquids: technology and applications *J. Mod. Opt.* **50** 539
- [2] Bonacic-Koutechy V and Mitric R 2005 Theoretical exploration of ultrafast dynamics in atomic clusters: analysis and control *Chem. Rev.* **105** 11–65
- [3] Judson R S and Rabitz H 1992 Teaching lasers to control molecules *Phys. Rev. Lett.* **68** 1500
- [4] Rabitz H, Vivie-Riedle R de, Motzkus M and Kompa K 2000 Whither the future of controlling quantum phenomena? *Science* **288** 824–8
- [5] Herek J L, Wohlleben W, Cogdell R J, Zeidler D and Motzkus M 2004 Quantum control of energy flow in light harvesting *Nature* **417** 533
- [6] Rabitz H, Hsieh M and Rosenthal C 2004 Quantum optimally controlled transition landscapes *Science* **303** 1998–2001
- [7] Wollenhaupt M, Prakelt A, Sarpe-Tudoran C, Liese D and Baumert T 2005 Quantum control and quantum control landscapes using intense shaped femtosecond pulses *J. Mod. Opt.* **52** 2187–95
- [8] Hsieh M, Wu R and Rabitz H 2007 The topology of the optimal control landscape for quantum ensemble preparation *J. Chem. Phys.* submitted
- [9] Tak-San Ho and Rabitz H 2006 Why do effective quantum controls appear easy to find? *J. Photochem. Photobiol. A* **180** 226–40
- [10] Rabitz H, Hsieh M and Rosenthal C 2005 The landscape for optimal control of quantum-mechanical unitary transformations *Phys. Rev. A* **72** 52337

- [11] Glaser S J, Schulte-Herbruggen T, Sieveking M, Schedletsky O, Nielsen N C, Sorensen O W and Griesinger C 1998 Unitary control in quantum ensembles: maximizing signal intensity in coherent spectroscopy *Science* **280** 421–4
- [12] Bonnard B and Chyba M 2003 Singular trajectories and their role in control theory *Mathematiques and Applications* vol 40 (Berlin: Springer)
- [13] Wu R, Pechen A, Rabitz H, Hsieh M and Tsou B 2007 Control landscapes for observable preparation with open quantum systems *J. Math. Phys. Preprint* 0708.2119
- [14] Helmke U and Moore J B 1994 *Optimization and Dynamical Systems* (Berlin: Springer)
- [15] Bender E A 1974 The asymptotic number of non-negative integer matrices with given row and column sums *Discrete Math.* **10** 217–23
- [16] Gail M and Mantel N 1977 Counting the number of  $r \times c$  contingency tables with fixed margins *J. Amer. Statist. Assoc.* **72** 859–62