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Exploring the impact of constraints in quantum optimal control through a kinematic formulation

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ABSTRACT

The control of quantum dynamics with tailored laser fields is finding growing experimental success. In practice, experiments will be subject to constraints on the controls that may prevent full optimization of the objective. A framework is presented for systematically investigating the impact of constraints in quantum optimal control simulations using a two-stage process starting with simple time-independent kinematic controls, which act as stand-ins for the traditional dynamic controls. The objective is a state-to-state transition probability, and constraints are introduced by restricting the kinematic control variables during optimization. As a second stage, the means to map from kinematic to dynamic controls is presented, thus enabling a simplified overall procedure for exploring how limited resources affect the ability to optimize the objective. A demonstration of the impact of imposing several types of kinematic constraints is investigated, thereby offering insight into constrained quantum controls.

1. Introduction

The control of quantum dynamics phenomena is an active area of theoretical and experimental research. Advances in femtosecond laser pulse-shaping, computer learning algorithms, and detection techniques have permitted increasing numbers of successful control experiments (for a review, see [1]). The experiments often seek to find an optimally shaped laser pulse that yields a high fidelity value for an objective. This paper concerns simulations that distinguish between quantum control in the traditional dynamical setting and a simplified analogous kinematic formulation described later. In a dynamic framework, the control field $\varepsilon(t),\ 0 \leqslant t \leqslant T$, is generally characterized by a large number of variables such as amplitudes and phases. In this paper a quantum system of N states is dynamically described by a diagonal field-free Hamiltonian H_0 and a field coupling transition dipole μ such that

$$H(t) = H_0 - \mu\varepsilon(t). \tag{1}$$

The dynamics are governed by the time-dependent Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}U(t,0) = H(t)U(t,0), \quad U(0,0) = \mathbb{1}, \tag{2}$$

with the formal solution

$$U(t,0) = \mathcal{T}\exp\left(-\frac{i}{\hbar}\int_0^t H(t')dt'\right), \tag{3}$$

where $\mathcal{T}$ is the time-ordering operator.

The search for an optimal control can be viewed as occurring over a quantum control landscape [2], which is defined as the objective as a function of the control variables. With dynamic control variables, each point on the landscape corresponds to a particular control field $\varepsilon(t),\ 0 \leqslant t \leqslant T$. Maximizing an observable entails climbing the landscape to its highest value by optimizing the controls. Here we consider the observable as the state-to-state transition probability at time T

$$P_{i\to f}[\varepsilon(t)] = |\langle f \mid U(T,0) \mid i\rangle|^2. \tag{4}$$

A critical point (i.e., an extremum) of the $P_{i\to f}[\varepsilon(t)]$ landscape corresponds to

$$\frac{\delta P_{i\to f}}{\delta\varepsilon(t)} = 0, \quad \forall\ 0 \leqslant t \leqslant T. \tag{5}$$

Evidence from carefully performed simulations shows that local (i.e., gradient-based) searches generally do not become stuck at suboptimal traps while climbing the $P_{i\to f}$ landscape [3]. This attractive behavior can be explained by the landscape being devoid of suboptimal critical points upon satisfaction of certain Assumptions [2]. Assumption (i) is that the quantum system is controllable [4,5], and (ii) is that $\delta U(T,0)/\delta\varepsilon(t)$ is full rank, where (ii) implies that a differential change $\delta U(T,0)$ has a corresponding variation $\delta\varepsilon(t)$ producing it. Naturally, these two Assumptions may be violated, but

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they appear to be easily satisfied for many quantum systems [3]. Importantly, Assumption (iii) is that the control field resources are unconstrained, which is of prime importance. In practice, for example, laser pulses are always constrained over a limited bandwidth around some operational frequency [6], although techniques to shift the frequency and extend the bandwidth as needed are becoming more prevalent [7–9]. Notwithstanding that in some cases readily accessible control laser resources may be fully adequate for obtaining an acceptably high quality yield, a major challenge is to reveal the impact of constraints on the erstwhile very attractive landscape behavior. Some works have addressed the use of constrained control resources through the inclusion of field fluence and/or spectral component restrictions [10–12]. The control time interval is also a resource, and recent work considered restricting T as a means to reduce the effects of decoherence caused by system-environment coupling [13–16]; however, there is a lower limit for T below which high objective fidelity can no longer be attained. Another study [17] used constrained numbers of pulse phases as control variables to examine the impact on state-to-state population transfer reflected in the altered landscape structure. In particular, when an inadequate number of phase parameters were used as the control variables, suboptimal traps and saddle regions appeared, which are absent from unconstrained state-to-state transition probability landscapes [18].

A general understanding of the impact of constraining the dynamic control variables upon the observable yield is extremely difficult to obtain due to the complex dependence of $U(T,0)$ in Eq. (3) on the controls. As a means to simplify the analysis of control landscapes and their critical point character, a kinematic formulation may be employed based on the identity

$$U = \exp(\imath A), \tag{6}$$

where A is an $N \times N$ Hermitian matrix such that

$$P_{i\to f}[A] = |\langle f \mid U \mid i\rangle|^2 \tag{7}$$

$$= |\langle f \mid \exp(\imath A) \mid i\rangle|^2. \tag{8}$$

The time-independent matrix A effectively subsumes the time-dependent Hamiltonian through an imposed equality of Eqs. (3) and (6)

$$\mathcal{T}\exp\left(-\frac{\imath}{\hbar}\int_0^T H(t)dt\right) = \exp(\imath A). \tag{9}$$

Eq. (5) can be expressed in terms of the matrix elements of A as

$$\frac{\delta P_{i\to f}}{\delta\varepsilon(t)} = \sum_{j,k}\frac{\partial P_{i\to f}}{\partial A_{jk}}\frac{\delta A_{jk}}{\delta\varepsilon(t)} = 0. \tag{10}$$

Upon satisfaction of the control landscape Assumptions, the set of functions $\delta A_{jk}/\delta\varepsilon(t)$ should be linearly independent over $0 \leqslant t \leqslant T$ [1,2]. In this case, a detailed analysis shows that Eq. (10) is satisfied only when $P_{i\to f} = 0$ and 1, which corresponds to null and perfect control, respectively [2]. The kinematic perspective utilizing the matrix A can be linked to the dynamic formulation through Eq. (9), and in the weak coupling limit A may be identified directly as the lowest-order term in the Magnus expansion [19]. In the kinematic formulation the time-independent elements of A act as the 'control variables'. The initial studies of control landscapes [2] exploited the ease of analysis using the kinematic perspective, and here we will follow this route to enable assessing the impact of control constraints.

The kinematic variables form a convenient set of stand-in controls which may be beneficially utilized in numerical simulations [20]. A transformation may then be made $A \to \varepsilon(t)$ to identify a dynamic control consistent with the kinematic control A. Concomitantly, the opposite transformation $\varepsilon(t) \to A$ may be readily performed. The dynamic formulation may also be generalized to consider all components of the full Hamiltonian (i.e., H_0, μ, and $\varepsilon(t)$ in Eq. (1)) as controls. Treatment of H_0 and μ as part of the controls naturally arises in varying the material or molecular character of the system [21] as well as when considering an imposed dynamic symmetry that may restrict the system's evolution [22]. Fig. 1 indicates the dual maps for transforming between dynamic and kinematic controls, $d \leftrightarrow A$.

There is considerable freedom and various means for performing the $d \leftrightarrow A$ transformations. An application-specific measure $\mathcal{L}$ that depends on d or A needs to be deliberately chosen according to the direction of the transformation. Natural choices for $\mathcal{L}$ include $P_{i\to f}$ or the full matrix U. Such choices for $\mathcal{L}$ specify what is preserved upon the transformation between kinematic and dynamic variables. A transformation from $A \to d$ can be specified through

$$J_{A\to d} = \min_d \|\mathcal{L}_{kin}(A) - \mathcal{L}_{dyn}(d)\|^2, \tag{11}$$

where $\mathcal{L}_{kin}(A)$ is first obtained using a kinematic control A and $\mathcal{L}_{dyn}(d)$ is dependent upon the chosen dynamic variables d. The minimization in Eq. (11) aims to find at least one set of dynamic controls d consistent with the kinematic controls as reflected through $\mathcal{L}$. Similarly, execution of a $d \to A$ transformation can be done through

$$J_{d\to A} = \min_A \|\mathcal{L}_{kin}(A) - \mathcal{L}_{dyn}(d)\|^2. \tag{12}$$

The transformations $A \to d$ and $d \to A$ may not be unique, depending upon the nature of $\mathcal{L}$ and especially the variables employed; furthermore, an A matrix with arbitrary integer multiples of 2π added to its eigenvalues will produce the same matrix U. A particular example for the transformation is specified by $\mathcal{L}_{kin} = U[A]$ and $\mathcal{L}_{dyn} = U[d]$, where $d = \varepsilon(t)$; if H_0 and μ are properly defined, there will generally be an infinite number of fields that can produce $J_{A\to d} = 0$. The non-unique nature of the kinematic → dynamic transformation implies that there can be a family of physically consistent controls, including in the case where kinematic constraints are imposed. It is important when transforming from constrained kinematic controls to their dynamic counterpart that no significant additional constraints be imposed on the dynamic controls in order to properly reflect the kinematic constraints. In this fashion, a constrained kinematic control can be mapped to the constrained dynamic controls. The present work is motivated by the consideration that the impact of constraints in A upon $\mathcal{L}$ is far easier to explore than directly addressing constraints in the vast space of dynamic control fields.

A main goal of this work is to present a systematic approach to investigate the impact of constraints while attempting to achieve the best possible control yield. As a first step, we will utilize kinematic controls with the knowledge that the results can be transformed when desired to the traditional dynamic scenario. A full

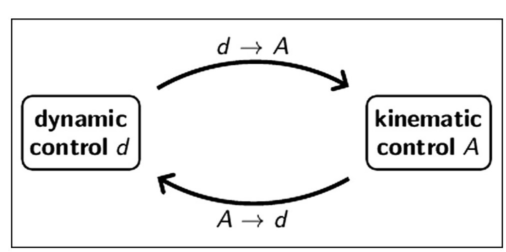

Fig. 1. Schematic of the transformation between dynamic controls and kinematic controls. The dynamic controls d may include an applied field and/or time-independent elements of the Hamiltonian; kinematic controls are treated as elements of a time-independent matrix A. The transformations are performed by specifying a measure $\mathcal{L}$ that can be compared as a means to identify A or d as appropriate (see Eq. (11) and (12) and the text for details). The present work illustrates the mapping from kinematic to dynamic controls in Section 4 with the choice $\mathcal{L} = U$.