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Some mathematical and algorithmic challenges in the control of quantum dynamics phenomena

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The theory and practice of control over quantum mechanical phenomena is receiving increasing attention, underscored by striking experimental successes. Nevertheless, many questions of fundamental and practical relevance to the field remain unresolved. With the aim of stimulating further development, this paper formulates a number of theoretical questions, divided into three categories. First, questions related to control law design are discussed, with an emphasis on controllability and optimal control theory. This leads to the second category of open problems relevant to closed loop laboratory implementation of quantum control, including learning and feedback methods. The sensitive dependence of control on basic quantum mechanical interactions motivates the third section, which treats coherent dynamical techniques for identifying the system Hamiltonian. An open issue overarching all of these directions is the need to discover general rules for the control of quantum systems. Although the list of issues raised in this paper is extensive, it should be viewed not as a complete menu for exploration, but rather as a springboard to new challenges as the field evolves.

KEY WORDS: quantum control theory, quantum dynamics, inverse problems

1. Introduction

Quantum mechanical control of molecular and other materials is a modern expression of the long-standing goal of directing the behavior of events at the atomic scale. What distinguishes quantum control from the traditional means of chemical manipulation is the use of delicate quantum wave interferences to alter the outcome of molecular scale dynamics phenomena. The products of this directed evolution are the prescribed control objectives (e.g., selective dissociation of a polyatomic molecule, the tailored manipulation of electron wavepackets in semiconductors, etc.). Recent progress in quantum control is highlighted by experimental successes in cleaving [1–3] and rearranging [3] selected chemical bonds, the creation of special excitations [4], the control of fluorescence in polyatomic molecules [5,6], and enhancement of radiative emission in high harmonic generation [7]. Control at the quantum level is the ultimate limit of materials manipulation, and many of the fundamental issues and implications of operating in this regime remain unresolved. This paper aims to stimulate development along these

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lines by posing some of the central questions that are evident from the present state of the field. In this context, selected areas of quantum mechanical control theory will be reviewed; however, this article is not intended to serve as a review of the field, for which the reader is referred to other works (cf. [8–10]).

Quantum control is most often expressed in terms of coherent motion stimulated by precisely tailored radiation interacting with an atomic or molecular system [11]. While this type of influence became possible with the advent of the laser in the 1960s, early attempts to achieve quantum control were met with some frustration [12–14]. Difficulties with the original experiments were attributed to features inherent in the underlying molecular dynamics, such as rapid intramolecular dissipation of energy. Although these are substantial concerns, in the late 1980s it was apparent that the main difficulties did not lie with the quantum systems themselves but rather with the design of the laser radiation: at the time, the design process depended primarily on physical intuition based on spectral information about the isolated molecules of the system. An important step was the recognition of the central role that quantum interference plays in the control process [15]. It became increasingly evident that the complex interplay between the control fields and the entities being controlled calls for the introduction of rigorous design tools. The required theoretical framework came with the realization that engineering control concepts can be extended to the quantum regime [16,17]. Coincidentally, the capability became available to shape laser pulses in accord with the demands of complex multi-frequency quantum dynamics. These developments have led to the recent dramatic progress in the experimental and theoretical investigation of quantum control, but many mathematical and algorithmic questions remain open.

The first step toward stating these questions is the mathematical expression of the quantum control problem. Most of this paper will work under the assumption that the system to be controlled can be characterized by its state function $\psi(t)$. This is a proper representation for isolated systems starting in a pure state; the complementary case arises, for example, in collisional or condensed regimes, when a density operator $\rho(t)$ must be introduced to describe the statistical mixture of states making up the system. The density operator formulation will be discussed where relevant in this paper.

Consider a quantum system that evolves from the initial state $\psi(t=0) \equiv \psi_0$. The objective of quantum control can generally be expressed as the desire for the evolving system to attain the set of expectation values

$$\widetilde{O}_j(t) = \langle \psi(t) | O_j | \psi(t) \rangle \tag{1}$$

for a specified collection of operators O_j, $j = 1, \ldots, N_O$. For each of these operators O_j, a set of times $\mathcal{T}_j = \{\tau_j^l; l = 1, 2, \ldots, L_j\}$ is defined, corresponding to the L_j instances or durations τ_j^l at which the expectation values of O_j are to attain (as best as possible) the control objectives $\widetilde{O}_j(t)$. Depending on the physical objective, an element $\tau_j^l \in \mathcal{T}_j$ may correspond to a discrete "target" time, a finite interval, or an infinite interval. Defining the set $\mathcal{T} = \bigcup_j \mathcal{T}_j$, the interval of control interest $[0, T]$ may be formalized as the smallest interval containing 0 and $\mathcal{T}$; below, we will assume that T is finite.

In the absence of any external control influence, evolution of the state function $\psi(t)$ under the Schrödinger equation is determined by the free Hamiltonian H_0, which by assumption does not yield dynamics producing the desired expectation values (1) in $[0, T]$. Quantum control theory considers the addition of a laboratory accessible *control law* term $C(t)$ to the Hamiltonian in order to achieve these objectives:

$$H = H_0 + C(t) \tag{2}$$

so that

$$\mathrm{i}\hbar \frac{\partial \psi(t)}{\partial t} = \big[H_0 + C(t)\big]\psi(t) \tag{3}$$

is the equation of motion.

The control law must belong to the relevant input space:

Definition 1. The input space $\mathcal{C}$ of a quantum control problem is the set of admissible maps $C : [0, T] \to A(\mathcal{H})$, where $A(\mathcal{H})$ is the space of admissible operators $\mathcal{H} \to \mathcal{H}$. Here, admissible implies satisfaction of regularity and other auxiliary conditions as well as consistency with the underlying physical problem.

With some abuse of notation, $C(t)$ will be written for the element C to emphasize time-dependence. Typically, $\mathcal{C}$ is expressed as a space of products of integrable, time-dependent, vector or scalar-valued functions with a fixed time-independent operator on $\mathcal{H}$. For example, a common input space law for lasers has the form $\mathcal{C} = \{-\boldsymbol{\mu}\cdot\boldsymbol{\varepsilon}(t)$: $\varepsilon_i(t) \in L^2[0, T],\ i = 1, 2, 3\}$, where $\boldsymbol{\mu}$ is the electric dipole operator, $\boldsymbol{\varepsilon}(t)$ is the applied electric field, and the index i refers to spatial orientation. Additional admissibility conditions may ensure that $\boldsymbol{\varepsilon}(t)$ obeys laboratory limitations on the range of achievable laser frequencies, intensities, energy, or other criteria.

In some applications, additional possibilities for $C(t)$ arise. These include (1) the use of magnetic fields, in which case the control law becomes $-\boldsymbol{\mu}_{\mathrm{m}} \cdot \boldsymbol{B}(t)$, where $\boldsymbol{\mu}_{\mathrm{m}}$ is the magnetic dipole operator and $\boldsymbol{B}(t)$ is the magnetic field, and (2) the use of materials whose design specifications themselves take the form of a control law, such as for quantum electron transport in semiconductors with variable material composition considered as the control. Here, however, we will confine the discussion to time-dependent controls based on an external electric field $\boldsymbol{\varepsilon}(t)$ coupled to the system through a dipole $\boldsymbol{\mu}$.

In some cases, it may be possible to obtain an adequate control description by replacing the Schrödinger equation (3) with a classical representation of the system dynamics. This is especially true for interatomic phenomena, because the de Broglie wavelength associated with atoms is often short relative to interatomic length scales. While the relationships between classical and quantum models of molecular evolution have been extensively investigated (cf. [18–21]), the implications of these relationships for control are not completely understood and will be addressed later in the context of the *quantum character* of the control problem.

Assuming knowledge of H_0 and a well-defined control law $C(t)$, equation (3) or its classical equivalent represents a complete model of the system of interest. If $C(t)$

is given *a priori*, the solution of equation (3) is a standard numerical problem in time-dependent quantum mechanics. However, the essence of the control problem is to find $C(t)$ such that the objectives in equation (1) are met, and since at least one of the control objectives lies in the future for any $t \in [0, T)$, this task presents some additional challenges. In particular, the Hamiltonian depends on the future state of the system through the control objectives, as can be formally represented by the expression $C(t) = C(\psi(s)\colon\ s \in [t, T])$. This non-causality introduces an entirely new set of mathematical issues which are not present in standard quantum or classical dynamics but are inherent to the theory and practice of temporal control in engineering and mathematical systems theory (see [22, and references therein] and [23–26]). Their implications for the quantum regime are central to the questions in this paper.

The formulation above is summarized in the following definition:

Definition 2. The quantum control problem consists of determining a control law $C(t)$ that causes the system to optimally achieve the expectation values (1) while possibly also satisfying auxiliary conditions. Quantum control theory encompasses methods of determining these control laws, their general properties, and their relationship to the underlying physical system and evolving quantum states.

The control of quantum phenomena involves a wide range of considerations and applications, and accordingly the balance of the paper will proceed with the overall structure shown in figure 1. Section 2 first addresses the fundamental question of the existence (often under prescribed auxiliary conditions) of a control law that causes exact satisfaction of the objectives (1). A general method for finding an optimal $C(t)$ is through minimization of a cost functional which penalizes deviations from the control objectives and auxiliary conditions; questions related to this quantum optimal control design are also discussed in section 2. Section 3 on the laboratory achievement of control addresses two limitations on the application of computational optimal control design to physical systems: (i) uncertainties in representations of H_0, $C(t)$, and (ii) the often-excessive computational demands of numerically solving the design equations. The techniques discussed in this section avoid these problems because they are directly based on the relationship between the actual observed physical system and the control law. In this context quantum control design can be seen as exploring the dynamical outcomes caused by $C(t)$ and providing estimates for the control law that may be refined in closed loop laboratory experiments. From another perspective, observations of these control processes might be a rich source of tailored data for identifying the system Hamiltonian and optical or other coupling terms. This is the subject of the section 4, which includes the development of inverse algorithms and the important possibility of realizing a new type of adaptive dynamical spectrometer. Section 5 covers the challenge of identifying control pathways and relationships between control laws for families of systems and objectives, including measures of similarity relevant to characterizing distinct quantum control systems. Identification of such rules for controlling quantum systems would be of central importance to the entire subject.

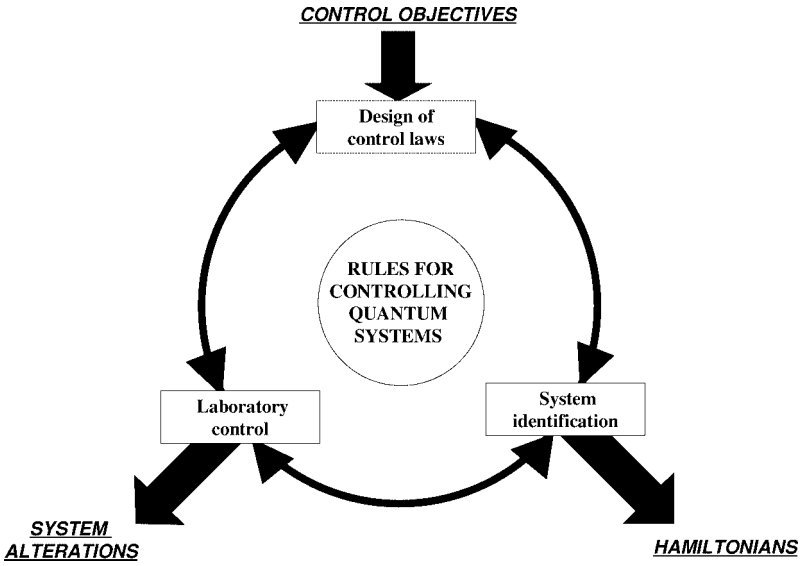

Figure 1. The relationships of the major quantum control topics discussed in this paper. The output of these efforts may be either alterations of an atomic or molecular system through its controlled manipulation, or enhanced fundamental understanding including identification of the system Hamiltonian. An overarching issue in all of these activities is the determination of the systematic rules for coherently manipulating quantum dynamics phenomena.

Statements of open problems are sequentially numbered throughout the paper, and are expressed as questions. Some of these questions may best be approached through extensions or applications of engineering control and mathematical systems theory to the quantum domain, while others may require the introduction of new methods. Whatever form the answers might take, we hope that the insight gained in considering these and related questions will contribute to the further development of the field: to quote a similar effort [27],

> "It is therefore entirely acceptable to treat these open problems in the way problems in this field have been treated in the past. After reformulation, simplification and modification, the open problem leads to a solution of a different problem that is perhaps easier but that is perhaps also more important."

We also emphasize that the collection of questions in this paper reflects the perspective of the authors, and that the list, expanded by those working in the field, will surely continue to evolve.

2. Design of control laws

The topics in this section concern the theory and practice of computationally designing the control law $C(t)$ in equation (3). Before we discuss the computations, it is natural to ask if the problem is well-posed such that a control law exists which will cause

the objectives and auxiliary conditions to be precisely satisfied (section 2.1). Even if the answer to the latter question is negative, one may still be satisfied with achieving the control objectives as best as possible through the optimization techniques of section 2.2.

2.1. Controllability of quantum mechanical systems

The fundamental importance of addressing controllability has long been recognized in engineering control applications; the broad literature on the classical aspects of the subject includes many comprehensive texts which cover linear [23,24] and nonlinear [22,25,26] controllability. In addition, several works have considered various aspects of quantum controllability, e.g., [8,28–35]; here, we will discuss some of the unresolved issues.

Quantum controllability is generally expressed in terms of identifying the set of final states that can be obtained from a given set of initial states. We first formalize some key notions by considering the infinite-dimensional quantum system prescribed by γ spatial variables in equation (3):

Definition 3. A state or wave function ψ is an element of the (complex) unit sphere $S = \{\psi \in L^2(\mathbb{R}^\gamma)\colon \|\psi\|_{L^2(\mathbb{R}^\gamma)} = 1\}$.

Solutions to Schrödinger's equation (3) define a trajectory $\psi(t\colon t \in [0, T])$ on S from the initial state $\psi(0)$; as such, $\psi(t)$ should have spatial derivatives of up to order 2 defined in (at least) the weak [36] sense. This motivates considering a subset of S with this (or a stronger) regularity property. For example, we may restrict our attention to $\psi \in (S \cap \mathcal{H}^2(\mathbb{R}^\gamma))$, where

Definition 4. The Sobolev space $\mathcal{H}^2(\mathbb{R}^\gamma) \subset L^2(\mathbb{R}^\gamma)$ contains all $\psi \subset L^2(\mathbb{R}^\gamma)$ such that ψ possesses weak (spatial) derivatives of up to order 2 belonging to $L^2(\mathbb{R}^\gamma)$.

We now introduce

Definition 5. The reachable set from ψ^1 is the set of states $\Psi(\psi^1) = \{\psi^2 \big| \exists C \in \mathcal{C}$ s.t. $\psi(t = t') = \psi^2\}$, where $\psi(t)$ satisfies (3) with $\psi(0) = \psi^1$ and t' is some finite time depending on ψ^1 and ψ^2.

Definition 6. The system (3) is controllable if $\Psi(\psi^1) = S\ \forall \psi^1 \in S$. In other words, the system is controllable if for any two states ψ^1 and ψ^2 there exists a control law $C(t\colon t \in [0, t']) \in \mathcal{C}$ such that $\psi(t = t') = \psi^2$, where $\psi(t)$ satisfies equation (3) with $\psi(0) = \psi^1$.

Truncating an infinite-dimensional quantum control problem with states in $\mathcal{H}$ to an n-dimensional problem (with states in $\mathcal{H}_n$) changes the nature of both the control and Hamiltonian operators and the states available as candidate members of reachable sets.

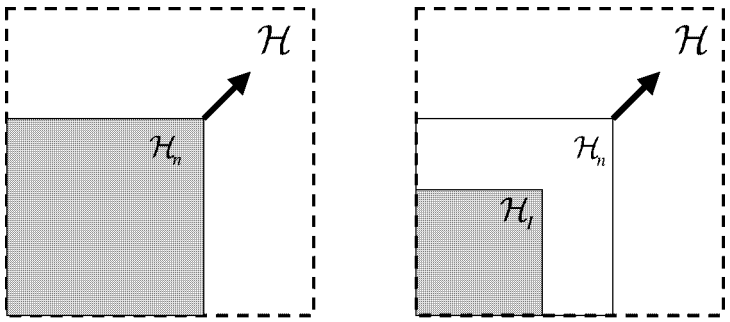

Figure 2. Pictorial representation of the controllability questions 1 (a) and 2 (b). In both cases, controllability is of interest within the shaded subspace as the dimension of the truncated space $\mathcal{H}_n$ increases to infinity.

This truncation in turn affects the controllability criteria given in definition 6. The concern is to characterize these effects by asking how a controllability result obtained in a finite-dimensional space relates to the original infinite dimensional problem from which it was derived; there are also inherently finite dimensional quantum systems (as with spins) where the latter consideration does not arise. The starting point is the result [31]:

Theorem 1. Let S be as in definition 3. Under general conditions (including boundedness of the control law), the set of unreachable states $S\backslash\Psi(\psi^1)$ from an arbitrary initial condition $\psi^1 \in \mathcal{H}^2(\mathbb{R}^\gamma) \cap S$ is dense in S. Furthermore, $(\mathcal{H}^2(\mathbb{R}^\gamma) \cap S)\backslash\Psi(\psi^1)$ is dense in $\mathcal{H}^2(\mathbb{R}^\gamma) \cap S$.

The theorem implies that within any open set around a point $\psi^2 \in \mathcal{H}^2(\mathbb{R}^\gamma) \cap S$ there exists a state unreachable from any $\psi^1 \in \mathcal{H}^2(\mathbb{R}^\gamma) \cap S$, and in addition that this unreachable state can be chosen to have weak derivatives in $L^2(\mathbb{R}^\gamma)$. Possible extensions of the theorem might show that, for example, $(C^2(\mathbb{R}^\gamma) \cap S)\backslash\Psi(\psi^1)$ is dense in $C^2(\mathbb{R}^\gamma) \cap S$ (thereby proving the existence of twice-continuously differentiable unreachable states).

Consider a quantum system that is controllable when its (truncated) equations of motion are expressed with respect to a particular n-dimensional basis which spans a finite dimensional space $\mathcal{H}_n$ when $n = n_0$. According to theorem 1, for every initial condition there must emerge a dense set of unreachable states in the limit n tends to infinity (depicted in figure 2(a)), assuming that the limiting process is well defined. In other words, once $n \to \infty$ (starting from $n = n_0$), the system must become uncontrollable in the strict sense defined above. This limit suggests the question:

Question 1. How is the controllability of a sequence of finite but increasingly higher dimensional quantum systems related to the controllability of the corresponding infinite-dimensional quantum system in the limit $n \to \infty$ (if this limiting process exists)? How are the sets of unreachable states that emerge in this limit characterized?

The analysis implied in question 1 can be subtle, as evident from a simple illustration involving the emergence or disappearance of unreachable states under finite increases in the dimensionality of $\mathcal{H}_n$. For example, in hydrogenic atoms the transitions due to emission or absorption of photons must satisfy the selection rules $\Delta l = \pm 1$ and $\Delta m = \pm 1$ or 0. If the step $\mathcal{H}_n \to \mathcal{H}_{n+1}$ of the limiting process adds a basis function to which there *does not* exist a sequence of allowed transitions from some function ψ^1 in $\mathcal{H}_n$, the additional dimension has caused a loss of system controllability. The converse situation may also arise where the additional basis function provides a "missing pathway" between states that were mutually unreachable in $\mathcal{H}_n$: in this case, the step $\mathcal{H}_n \to \mathcal{H}_{n+1}$ might cause an uncontrollable system to become controllable. It is an open question in quantum controllability to understand how such stepwise processes may be interpreted in the infinite limit, which must be taken with special care and in some cases may not exist.

The concept of controllability under increases in state space dimensionality may be related to general properties of function approximation. For example, consider the approximation of an arbitrary element f of an (infinite-dimensional) function space F by linear combinations $\{\sum_{i=1}^{n} a_i f_i\}$ of an n-dimensional basis $\{f_i\colon\ i = 1, \ldots, n\} \equiv F_n \subset F$. The n-term approximation is accurate if the coefficients a_i of the corresponding n_+-term expansions for f are negligible for $i > n$. The condition for extending controllability from an n- to an n_+-dimensional system can be viewed as the converse: for ψ^2 as in definition 6, each coefficient in the expansion $\psi^2 = \sum_{i=1}^{n_+} a_i \psi_i$ must be able to take any complex value (under the constraint $\sum_{i=1}^{n_+} |a_i|^2 = 1$).

Now consider the related issue of controllability within a "subspace of interest" $\mathcal{H}_I$ that is contained within $\mathcal{H}_n$ (i.e., as depicted in figure 2(b)). Let $\mathcal{H}_I$ be spanned by the first I elements of the set of basis functions $\{\psi_i\colon\ i = 1, \ldots, n\}$ spanning $\mathcal{H}_n$. Definition 6 may be modified to restrict analysis to the subspace of interest: controllability will be taken to mean that a system is controllable between any two states ψ_I^1 and ψ_I^2 in $S \cap \mathcal{H}_I$. Controllability may be described as stationary within $\mathcal{H}_I$ if it remains unchanged as individual dimensions are added in any order to $\mathcal{H}_n$ until (if it exists) the limit $\lim_{n\to\infty} \mathcal{H}_n = \mathcal{H}$, $n \geqslant I$, is obtained. This suggests the question:

Question 2. What characteristics of the Hamiltonian H_0, the dipole or other coupling coefficients, and the spaces $\mathcal{H}_I \subset \mathcal{H}_n \subset \mathcal{H}$ are required for stationary controllability within $\mathcal{H}_I$?

Questions 1 and 2 do not address the effects on the evolution of states within the truncated space $\mathcal{H}_n$ arising from states that lie outside of $\mathcal{H}_n$. This consideration also has practical consequences. For example, suppose that controllability is satisfied within $\mathcal{H}_n$ for question 1 or within $\mathcal{H}_I$ for some $\mathcal{H}_n$ in question 2. A realizable laboratory control might inadvertently also access states lying outside of $\mathcal{H}_n$ which might even lift the controllability in the desired subspace. Techniques from optimal control theory would be the desirable way to handle the discovery of practical fields best satisfying the assumptions under an associated controllability analysis.

Following upon the latter discussion, a new class of questions is introduced if a term is added to the Schrödinger equation to represent the interaction of the remainder states that are not explicitly modeled: elements of $\mathcal{H}_r \equiv \mathcal{H}\backslash\mathcal{H}_n$. One such term introduced in [37], cf. also [22, and references therein] is an n-dimensional disturbance vector w:

$$\frac{\partial\psi}{\partial t} = \big[H_0 + \mathrm{C}(t)\big]\psi + w, \tag{4}$$

where $\psi \in \mathcal{H}_n$. The form and magnitude of the disturbance term w is problem-dependent and assumed to be given *a priori*, and generally leads to a nonunitary evolution for $\psi(t)$. In min–max optimal control theory (cf. section 2.2), w is selected to maximize the disruptive effect of the energy-bounded disturbance (e.g., fluctuations in the laboratory environment and apparatus). In another general context, w could represent coupling to a bath external to the dynamics described by $H_0 + C(t)$. This type of coupling is important for considerations of dynamical cooling [38–41] in the analogous density matrix formulation.

Question 3. What general models of dynamics exterior to $\mathcal{H}_n \subset \mathcal{H}$ can cause controllable systems to become uncontrollable and vice versa? Also, how does introducing a disturbance affect answers to questions 1 and 2?

Lie group analysis [26] has been applied to determining quantum controllability, and has resulted in the identification of a sufficient condition for controllability in n-dimensional spaces $\mathcal{H}_n$ [30]. The analysis exploits the facts that states unit-normalized in some $\mathcal{H}_n$ lie on the $(2n-1)$-dimensional unit sphere, and that there exist $n \times n$ unitary transformations U (elements of the Lie group $U(n)$) that map between any two points on such a sphere. A Lie algebra of matrices is generated by the Lie brackets associated with the evolution operator of the Schrödinger equation (3), and a sufficient condition for controllability in $\mathcal{H}_n$ is that this algebra have dimension $n^2 - 1$. While this condition holds under the constraint that the control field amplitudes are bounded [30], an open question suggested in this work is the extension of the result to stricter (and more realistic) admissibility conditions:

Question 4. Can the Lie algebraic controllability conditions [30] be extended to treat the case where both the amplitude and the frequency of the control field are bounded from above and below?

This issue has practical significance as it prescribes real laboratory conditions.

The quantum control problem becomes more complex when the control law and the free Hamiltonian cannot be treated independently. An important example of this phenomenon is intense-field laser control of molecular motion, where the electric field

can directly alter the dipole operator through its manipulation of the electronic degrees of freedom:

$$C(t) = -\boldsymbol{\mu}\big(\boldsymbol{\varepsilon}(t)\big) \cdot \boldsymbol{\varepsilon}(t). \tag{5}$$

In simple cases, the relation in equation (5) may be expanded in terms of a low order polynomial in $\boldsymbol{\varepsilon}(t)$ whose coefficients are the electric moments and polarizabilities of the system. Of special interest are situations in which the nonlinear structure in (5) may affect the controllability of the system (including the positive case in which this interaction makes a previously inaccessible target reachable):

Question 5. Can the existing methodology for proving quantum controllability be adapted, or a new methodology from mathematical systems theory be applied, to treat the situation where the control law has a nonlinear dependence on the control field?

The circumstances motivating question 5 can also be viewed from the larger perspective of a controllability analysis simultaneously including electronic and nuclear motion. In the latter circumstance the control field will enter the Hamiltonian linearly, but at the expense of explicitly including the electronic degrees of freedom. When electronic excitation is under study and when the Born–Oppenheimer approximation is not valid this full analysis will be required. The same comment also applies to the performance of optimal control designs in the strong-field regime. The practical importance of investigating the latter domain has recently been demonstrated experimentally [3].

A complementary situation occurs when the back action of the quantum *medium* upon the propagating control field is significant (i.e., the medium is optically dense). This scenario has been examined experimentally for a vapor of sodium [42], and the topic is of practical importance because the controlled medium will be dense in any application directed toward collecting large amounts of product. Optically dense media can interact with the electric field to alter its phase and/or amplitude structure as it propagates. In order to model this effect, the Schrödinger equation must be coupled with Maxwell's equations:

Question 6. In what cases can controllability be shown within the product state space of the coupled Schrödinger–Maxwell equations for optically dense media?

In practical applications the medium will be at a finite temperature. Hence, to answer question 6 (or any other question that concerns a statistical mixture of quantum states) the density operator formulation is necessary. In this formulation, the time evolution is given by the quantum Louiville equation

$$\frac{\mathrm{d}\rho(t)}{\mathrm{d}t} = \frac{1}{\mathrm{i}\hbar}\big[H(t), \rho(t)\big], \tag{6}$$

and expectation values are calculated as

$$\big\langle O(t)\big\rangle = \mathrm{Tr}\big(\rho(t)\,O\big). \tag{7}$$

The relevant definition of controllability becomes:

Definition 7. The system (6) describing the evolution of the density operator $\rho(t)$ is *controllable* if for any two density operators ρ^1 and ρ^2 there exists a control law $C(t) \in \mathcal{C}$ such that given the initial condition $\rho(0) = \rho^1$, then $\rho(t') = \rho^2$ for some finite time t'.

While the Schrödinger–Maxwell system has a product state space representing both $\rho(t)$ and $\boldsymbol{\varepsilon}(t)$, expectation values (7) depend only on the density operator, which is the usual focus of controllability studies. This inspires the question:

Question 7. In what, if any, cases is it possible for the Schrödinger–Maxwell system to be controllable in the state space of the quantum state but not controllable in that of the electric field, or vice versa?

The latter case of controlling the electric field is of importance in the allied subject of optical field propagation, e.g., [43].

The next question seeks to address controllability from an analysis of the kinematic structure of the Hamiltonian. When expressed with respect to a finite (orthonormal) basis of eigenstates $\{\psi_i\}$, the Hamiltonian in equation (3) often takes the form:

$$H_0 = \begin{pmatrix} E_1 & \dots & 0 \\ & E_2 & \\ \vdots & \ddots & \\ 0 & & E_n \end{pmatrix}, \qquad C(t) = \begin{pmatrix} 0 & c_{12}(t) & \dots & c_{1n}(t) \\ c_{12}^*(t) & 0 & c_{ij}(t) & \vdots \\ \vdots & c_{ij}^*(t) & \ddots & \\ c_{1n}^*(t) & \dots & & 0 \end{pmatrix}. \tag{8}$$

In the case of optical excitation with an arbitrary electric field $\boldsymbol{\varepsilon}(t)$, the control law $C(t)$ matrix elements are $c_{ij}(t) = -\boldsymbol{\varepsilon}(t) \cdot \langle \psi_i | \boldsymbol{\mu} | \psi_j \rangle$. In the present analysis, we assume that the transition frequencies are incommensurate such that the control field can independently address each transition. This condition is $|E_i - E_j| \neq |E_k - E_l|$ for all values of the indices i, j, k, and l with the constraints $i \neq j$ and $k \neq l$ as well as at least one of the indices being distinct between the pairs $\{i, j\}$ and $\{k, l\}$. The connectivity amongst the states $\{\psi_i\}$ provided by the elements c_{ij} is central to issues of controllability. The structure in C can be conveniently expressed graphically by employing the following definitions:

Definition 8. Let every state be a node of a graph and let there be edges between every pair of nodes i and j with c_{ij} being nonzero. Then two states are connected by a path of length m if there exists a connected set of m edges between i and j.

Definition 9. The graph is *connected* if there exists a path of some length m between every pair of vertices. A natural upper bound on m is $n - 1$ (the longest nonredundant path).

Definition 10. The $n \times n$ adjancy matrix A has elements $a_{ij} = 1$ if $c_{ij} \neq 0$ and $a_{ij} = 0$ otherwise.

It follows from these definitions that $(A^2)_{ij} = \sum_k a_{ik}a_{kj}$ is nonzero if and only if a_{ik} and a_{kj} are both nonzero for at least one k, implying the existence of a path of length 2 through k from i to j. This is easily generalized for paths of length m: $(A)^m_{ij} = \sum_k (A)^{m-1}_{ik}(A)_{kj}$ is nonzero if and only if there is at least one path of length $m-1$ from i to some k, and a path of length 1 from this k to j. Observing that the elements of A^m are non-negative, it follows that a graph is *connected* if and only if $\sum_{m \leqslant (n-1)} A^m$ has no zero off-diagonal elements [43]; this implies the existence of a *kinematical* pathway from any initial state to any final state $\psi(t') = \sum_{i=1}^n a_i \psi_i$.

The conditions of connectivity and incommensurate transition frequencies involve only the eigenvalues of H_0 and powers of the adjacency matrix A (which depends only upon which elements of C are nonzero, rather than their particular values) and are thus fundamentally different from the Lie algebraic criteria for controllability found in [30]. An open question is whether these new conditions are sufficient for controllability:

Question 8. Does graph connectivity imply controllability of a finite-dimensional quantum system with incommensurate transition frequencies?

Progress toward answering this question has recently been made in [32,33].

The analysis of controllability in terms of state functions $\psi(t)$ reaches beyond what is necessary physically, as realistic objectives are expectation values of observable operators (cf. equation (1)). Since these quantities involve integrals of state functions, their control should generally be less demanding than that of the state itself. However, the quadratically nonlinear nature of the expectation values adds a level of additional complexity to the problem of determining controllability. These observations raise the question:

Question 9. Do there exist general conditions for the controllability of objective expectation values that relax those for the controllability of quantum states themselves?

In [31], this question is answered for the special case of projection operators $O_i = |\psi_i\rangle\langle\psi_i|$. In this situation, the control objectives are populations of quantum states (e.g., $|\langle\psi|\psi_i\rangle|^2$), and it is shown under general conditions that the controllability of these objectives can be established. Kinematical constraints on the controllability of systems in mixed states have been established [34,35], based on the eigenvalues of $\rho(0)$ and those of the objective operator.

The proof of controllability in [31] establishes an algorithm which *constructively* generates control laws for objective operators of the form $O_i = |\psi_i\rangle\langle\psi_i|$ in terms of sinusoidal electric fields of different fixed frequencies. This, along with other work on special cases of constructive quantum control [45,46], invites the question:

Question 10. Can general constructive control solutions be developed for the quantum controllability of a broad class of objective expectation values?

While the explicit construction of control solutions is generally an area of active research in control theory [26,47], the case of quantum controllability of expectation values reduces this task to the specific structure of Schrödinger's equation and may be amenable to attack.

A serious practical issue in implementing the control design process (cf. section 2.2) is the need for the accurate numerical solution of the Schrödinger equation (3), which can be computationally very expensive. Since the Schrödinger equation must be solved at least once (and generally many times) in most optimal control and Hamiltonian identification methods, the numerical evolution of quantum systems with high degrees of freedom *must* be approximated in some fashion before these techniques can be applied. Along these lines, broad classes of quantum dynamics approximations have been developed, and in principle any of them could be applied to quantum control design. Before actually attempting to attain designs, it is worth investigating the following question:

Question 11. What are the effects on controllability of replacing the Schrödinger equation with its various quantum dynamical approximations?

Significant influence of a dynamical approximation upon a system's controllability could have serious consequences for the reliability of any resultant control designs based on the approximation.

2.2. Optimal control theory

2.2.1. Basic concepts

As section 2.1 suggests (cf. question 10), the establishment of controllability generally does not provide an actual control law required to achieve an objective of the form (1); full controllability simply means that at least one such control law exists. Several approaches for determining control laws will be discussed in the remainder of this paper. The present section concerns the use of optimal control theory for this purpose. An extensive literature on optimal control theory can be found in classical engineering and mathematical systems theory (e.g., [22,48,49]) and increasingly in quantum mechanics (e.g., [8, chapter 6 and references therein] and [50,51]). Considering control law design as an optimization problem is quite natural, as attaining the best possible final level of control is always the goal; further, optimization is essential when there are competing physical objectives that must simultaneously be met.

In addition to the general approach outlined below, special techniques for control law design have been developed for cases in which *a priori* specification of control mechanisms is possible. These methods include time-resolved "pump-dump" or frequency-resolved multiple-path and simulated Raman adiabatic passage (STIRAP) schemes. Under particular quantum dynamics approximations and/or assumptions, these techniques

allow for the derivation of closed form expressions for control laws that optimally or exactly accomplish certain control objectives. The literature on related theoretical (see [8, chapters 3–5 and references therein] and [52–62]) and experimental (see [8, chapters 3–5 and references therein] and [59,60,63,64]) developments is extensive.

The first step in formulating the general quantum optimal control problem is to define a "cost functional" whose minimization represents the balanced achievement of control and possibly other objectives. This cost functional is given by

$$J\big(C(t), \psi(t); t \in [0, T]\big) = \sum_k J_k\big(C\big(\psi(t); t \in [0, T]\big)\big), \tag{9}$$

where each term is positive semi-definite and the goal is to minimize J with respect to $C(t)$. While the specific form of the cost functional is flexible and problem-dependent, a term J_1 that addresses the achievement of the optimal control objectives (1) is always included, such as:

$$J_1 = \sum_{j=1}^{N_O} \sum_{l=1}^{L_j} W_{1,j}^l \begin{cases} \displaystyle\int_{\tau_j^l} \mathrm{d}t \left|\langle O_j(t)\rangle - \widetilde{O}_j(t)\right|^2 & \text{if } \tau_j^l \text{ is an interval,} \\ \left|\langle O_j(\tau_j^l)\rangle - \widetilde{O}_j(\tau_j^l)\right|^2 & \text{if } \tau_j^l \text{ is a discrete time.} \end{cases} \tag{10}$$

Here, the $W_{1,j}^l$ are positive design weights assigned to each of the objectives; for full generality these weights could be time dependent and appear in the integrand in equation (10).

For physically realistic control laws, the energy of the laboratory/molecular interaction must be bounded. This criterion is often included by adding the term

$$J_2 = \int_0^T W_2(t) \left|\boldsymbol{\varepsilon}(t)\right|^2 \mathrm{d}t \tag{11}$$

to the cost functional, which effectively limits the total electric field fluence. Here, $W_2(\mathrm{t}) \geqslant 0$ determines the time-dependent relative importance of minimizing the fluence. Note that the term J_2 does not prevent $\boldsymbol{\varepsilon}(t)$ from being large in some small interval of time, although a cost on the local magnitude at any time could be introduced for this purpose.

Penalty terms may also be included, causing the minimization of the expectation values of $N_{O'}$ "undesirable" operators O_j' at the corresponding times $\tau_j'^l$:

$$J_3 = \sum_{j=1}^{N_{O'}} \sum_{l=1}^{L_{j'}} W_{3,j}^l \begin{cases} \displaystyle\int_{\tau_j'^l} \mathrm{d}t \left|\langle O_j'(t)\rangle\right|^2 & \text{if } \tau_j'^l \text{ is an interval,} \\ \left|\langle O_j'(\tau_j'^l)\rangle\right|^2 & \text{if } \tau_j'^l \text{ is a discrete time.} \end{cases} \tag{12}$$

In addition to those explicitly given here, there are many other forms of J_k that could be incorporated into the cost functional. These terms could represent, for example, restriction of the windowed Fourier transform of $\boldsymbol{\varepsilon}(t)$ to a particular frequency band, minimization of sensitivity to small perturbations in the control law (as will be discussed below), or other characteristics of the desired optimal control solution.

One property of control law solutions $C(t)$ of practical import is simplicity. Several measures of simplicity could be used, such as the ability to decompose the control law into only a small number of spectral components with high accuracy [65]. However, the notion of field simplicity is best associated with the ease of stable and reliable generation in the laboratory, rather than any preconceived sense of simplicity associated with the presence of few field components. Design of simple control laws might be accomplished by introducing a term in the cost functional that favors solutions with suitable characteristics, or in a very *ad hoc* fashion by starting an iterative optimization algorithm with a simple control field and halting the process while some of this simplicity is still preserved but likely before complete convergence to the control objectives has been achieved. The latter suggestion follows from the observation that the final small fraction of progress toward the control objectives is often responsible for most of the complexity in the control field [66], (e.g., see [8, table 6.1]). None of the above approaches has been subjected to a careful mathematical analysis, and further efforts to characterize the effects of these modifications on the optimal control process may be useful.

Thus far the terms in the cost functional have all been introduced to seek a control field that biases the objective or other goals in some specified direction. Under favorable circumstances one or more of these costs could be re-expressed as a hard demand by introducing a Lagrange multiplier. An example would be a requirement that the laser pulse energy be fixed at a specified laboratory accessible value; the reshaping would redistribute that energy as best as possible over a band of frequency components to meet the physical objective. Some absolute demands may lead to inconsistencies and resultant numerical design difficulties if the demand cannot be satisfied for some (often hidden) dynamical reason. Deducing when hard physical demands are attainable would be valuable, leading to the question:

Question 12. What characteristics of the Hamiltonian and the dipole operator can be used to identify attainable hard constraints when a system is put under control?

The question seems naturally suited for a controllability analysis.

Once the cost functional (9) has been defined, the optimal control law is determined by minimizing the cost functional over the function space of admissible controls. Local or global optimization algorithms (e.g., respectively, gradient descent and genetic algorithms) may be used to find the minimum of (9) subject to satisfaction of the Schrödinger equation, possibly under suitable quantum dynamics approximations and assumptions [67,68]. Alternatively, the Euler–Lagrange approach may be pursued, as explained below.

At the relevant minima of the cost functional, the first order variation with respect to the control law vanishes:

$$\frac{\delta J}{\delta C(t)} = 0. \tag{13}$$

Equation (13) is subject to the dynamical constraint that $\psi(t)$ satisfies the Schrödinger equation; this may be assured through the introduction of a Lagrange multiplier func-

tion $\lambda(t)$ [22]. The resultant variational problem produces Euler–Lagrange equations whose solutions define the controls $C(t)$ operative at each local extrema of J.

To demonstrate some of the characteristics of these equations, consider as an example a quantum optimal control problem in which there is only one objective operator O whose expectation value is to be optimized at the single target time T. The cost functional $J = J_1 + J_2$ consists of the terms given in equations (10) and (11), both weights are set to unity, and $C(t) = -\boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}(t)$. The Euler–Lagrange equations then become:

$$\mathrm{i}\hbar \frac{\partial \psi(t)}{\partial t} = \big[H_0 - \boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}(t)\big]\psi(t), \quad \psi(0) = \psi_0, \tag{14}$$

$$\mathrm{i}\hbar \frac{\partial \lambda(t)}{\partial t} = \big[H_0 - \boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}(t)\big]\lambda(t), \quad \lambda(T) = 2\big(\langle \psi(T) | O | \psi(T) \rangle - \widetilde{O}\big) O \psi(T), \tag{15}$$

$$\varepsilon_i(t) = -\frac{1}{\hbar} \operatorname{Im}\big\{\langle \lambda(t) | \mu_i | \psi(t) \rangle\big\}. \tag{16}$$

Here, the subscript i refers to spatial orientation. If the field in equation (16) is substituted into equations (14) and (15), the system becomes a pair of coupled nonlinear evolution equations. The initial and final conditions for equations (14) and (15) are $\psi(0)$ and $\lambda(T)$, and they generally require an iterative solution to the equations. Henceforth, equations of the form (14)–(16) will be referred to as the quantum optimal control equations.

Numerical experiments [69, figures 15 and 16] have shown that $|\lambda(\mathbf{x}, t)|^2$ may be quite similar in form to $|\psi(\mathbf{x}, t)|^2$. This suggests asking whether suitable physical interpretations of the Lagrange multiplier function may aid in attaining desirable control behavior:

Question 13. Considering that the quantum optimal control problem is generally underposed, to what degree may the Lagrange multiplier function be exploited to obtain a control solution with desired physical characteristics in the evolution toward the control objectives? Can the similarity of $|\lambda(\mathbf{x}, t)|^2$ and $|\psi(\mathbf{x}, t)|^2$ be utilized to produce viable approximate solutions to the optimal control equations?

Systems of the form (14)–(16), as two-point boundary value problems in time, may have any number of solutions. Under a mild set of assumptions the general quantum optimal control problem has been shown to possess a countable infinity of solutions [70]. This result has been shown for cost functionals of the form $J = J_1 + J_2 + J_3$ having one objective operator O at a final time T and a single penalty operator O' evaluated over the entire control interval (i.e., $\mathcal{T}' = \{[0, T]\}$). In this work [70] additional assumptions are that: (i) O and O' are bounded operators, (ii) O is either positive- or negative-definite, and (iii) $\boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}(t\colon\ t \in [0, T])$ is bounded (although the proof can be extended for unbounded control terms). However, it is known that at least some quantum optimal control problems have a unique solution, as was demonstrated in [17] for the problem of

a harmonic oscillator system with a particular quadratic cost functional. These overall observations lead to the question:

Question 14. What special criteria existing in the Hamiltonian structure or imposed on a quantum optimal control problem will permit the existence of a unique solution?

As analytical solutions to the quantum optimal control equations cannot generally be found, iterative numerical algorithms must be employed. The time-dependent Schrödinger equation in multiple spatial variables is computationally very expensive to solve, raising the question:

Question 15. Can quantum dynamics approximations can be found that are tailored to efficiently solving the quantum optimal control equations?

There appear to be many opportunities to develop special control design approximations, and while some work has already been done [8], there is room for much development.

An algorithm was developed in [71,72] that converges to a local solution of the quantum optimal control equations monotonically and at a quadratic rate. This method has been extended for use in the density matrix framework [73], and is applicable to cost functions depending on a single-time cost of the form $J\{\psi(T), \{O_a\}\}$, where J has a positive-semidefinite Hessian $\delta^2 J/(\delta\psi(t)\delta\psi(t'))$ and $\{O_a\}$ is some set of objective operators. These developments invite extension:

Question 16. Can a monotonically convergent algorithm be found to solve the quantum optimal control equations for arbitrary objectives? Can algorithms be found that converge faster than quadratically?

A large body of numerical studies provides examples of solutions to the quantum optimal control equations. However, none of this work has illuminated the *general* behavior, stability, and classes of solutions to the quantum optimal control equations (14)–(16) (here, by classes of solutions we mean the qualitative notion of groups of solutions with particular properties, such as nondispersivity, periodicity, etc.). Furthermore, although there exists a broad literature on the subject of nonlinear Schrödinger equations (NLS's), e.g., [74–76], this mathematical analysis has not explicitly been extended to the allied quantum optimal control equations. This suggests the following question:

Question 17. What mathematical techniques can be applied to the quantum optimal control equations to reveal the classes of behavior admitted by their solutions?

The possibility that unusual behavior can be expected is evident from one study which showed that the quantum control equations can be made equivalent to the standard NLS under suitable conditions [77] (see also section 3.3).

The topics and questions raised to this point have not explicitly treated the impact of random environmental influences and the finite precision of the laboratory apparatus on the evolution of controlled quantum systems. In general, noise in $C(t)$ is thought of as harmful in the context of trying to achieve control objectives. However, hints from the subject of stochastic resonance [78] suggest that under suitable conditions noise may possibly have beneficial effects, such as allowing the achievement of a particular level of control using smaller total field fluence than that required for the noise-free system. It remains an open problem to answer:

Question 18. Under what, if any, conditions can the presence of noise assist in the achievement of quantum control objectives? What are the possible physical mechanisms behind these effects?

The next section will return to the standard view of environmental influences, fluctuations in the laboratory apparatus, and other uncertainties as being undesirable. The focus will be on methods for understanding and minimizing their effects on quantum control systems.

2.2.2. Robust designs

Due to imperfect knowledge of system Hamiltonians and coupling operators as well as the limited precision and presence of background fluctuations inherent to any laboratory apparatus, it is impossible to perfectly reproduce either optimally designed control laws or the exact specifications under which they were designed. Hence, it is important to study the sensitivity of the control objective or cost functional to random variations or uncertainties in the operators and initial conditions describing the evolution of the system. There is extensive work on the general topic of robust optimal control in the engineering [79–81] and quantum control [82,84,86] literatures.

A general approach to assessing robustness and stability in quantum control has been considered [82] based on introducing a stability operator S, the kernel of which is related to the curvature $\delta^2 J[\varepsilon]/\delta\varepsilon(t)\delta\varepsilon(t')$ of the cost functional with respect to the control law. Considering the curvature is necessary as the null value of the first order variation $\delta J[\varepsilon]/\delta\varepsilon(t) = 0$ defines the optimal solution. Conditions for robustness and optimality of the control solutions can be expressed in terms of the spectrum of S, and this analysis can also reveal qualitative relationships between the various terms in the cost functional and the robustness/optimality features of the control solutions. The open questions suggested by this analysis include questions 19 and 20:

Question 19. As the eigenvalues of the stability operator S are an important determinant of the stability and robustness of an optimal control solution, can the dominant characteristics of a system Hamiltonian, coupling operators, and cost functional be established that determine these eigenvalues?

Question 20. Will the classical and quantum formulations of the same molecular scale control problem exhibit distinctly different stability behavior?

The introduction of a penalty term of the form

$$J_3 = \int_0^T W_3 \left| \left\langle \psi(t) \right| O' \left| \psi(t) \right\rangle \right|^2 \mathrm{d}t, \tag{17}$$

where O' is an arbitrary positive definite operator, was observed [82] to improve the robustness of optimal control solutions. The presence of J_3 can bias the system to satisfy demands tangential to the true control objectives, causing an effective "drag" along the way to the goal. Hence, the effect of J_3 may be loosely interpreted as analogous to the presence of viscous drag in stabilizing a classical mechanical system about a weakly stable point in its phase space. However, the possible stabilization mechanisms have not been carefully studied or characterized, leaving the open question:

Question 21. Can a complete mechanism be put forth explaining how the introduction of suitable ancillary objectives may stabilize the solutions to quantum optimal control problems?

The robustness effects of penalty operators with more specific forms than that given by equation (17) may be easier to intuit. For example, the term

$$J_s = \int_0^T \mathrm{d}t \left(\frac{\delta \langle O(T) \rangle}{\delta \varepsilon(t)} \right)^2$$

(or analogous expressions with higher derivatives) may be used [83] to reduce the sensitivity of the achieved control objectives at the target time T to uncertainty in control fields. Analogs of this penalty term for the sensitivity of the target objective to uncertainty in other variables were found [84] to be capable of reducing the sensitivity to errors in force constants and other model parameters.

Design of robust quantum optimal control solutions can be achieved through the min–max procedure, which involves simultaneously maximizing the effects of an energy-bounded disturbance and minimizing the objective functional (9). Solutions to such min–max problems represent the best possible control under the worst possible energy-bounded disturbances. For linear dynamical systems the min–max problem becomes H_∞ control, which has an exact solution through the Ricatti equations. This procedure is well-developed in engineering control theory [85], and it has been applied to robust control designs for selective vibrational excitation in molecular harmonic oscillators [37]. In general, the min–max technique tends to give conservative robust solutions as it works against the worst possible bounded disturbance, but encountering this worst disturbance in practice is unlikely. This point suggests the following question for consideration of a less extreme class of disturbances:

Question 22. What are the robustness characteristics of solutions to a "mollified" min–max analysis, in which the disturbances are expressed by a distribution of the most likely fluctuations in the environment?

Explicit modeling of this or related circumstances may involve stochastic versions of the Schrödinger equation (3). The resulting analysis should give designs that are robust under more realistic conditions than those modeled in a worst case scenario.

The conclusions of min–max studies [86] reinforce the importance of question 22. For a diatomic molecule modeled as a Morse oscillator, the robustness properties of solutions to the min–max equations were compared with solutions to the standard Euler–Lagrange equations (cf. (14)–(16)) derived without any robustness considerations. While the min–max controls performed better under the application of the worst-possible disturbance (for which they were designed), they did *not* necessarily outperform the standard Euler–Lagrange solutions under disturbances other than the worst case. For example, min–max control fields were demonstrated to be significantly *less*-robust than standard Euler–Lagrange control fields to sinusoidal perturbations with the same amplitude constraints as the worst-case disturbance. This underscores the importance of designing control laws that are robust to the particular class of disturbances most likely to occur.

Even in cases where the robustness properties of two designs are quite distinct, simulations have shown [37] that robust control designs may differ only slightly from nonrobust designs (i.e., the L^2 norm of the difference between the two control laws may be small). This similarity suggests that robustness properties in some cases may result from very subtle effects. It was also noted [37] that the relationship between the robust field and the standard design (i.e., created without robustness considerations) can take two forms: the robust field can be either a scaled, self-similar version of the standard field (which may be described as achieving robustness by "speaking louder") or can have a qualitatively different form. At present, no means exists to predict in general when either of these two cases will occur, suggesting the question:

Question 23. Can features of optimal control problems be identified that predict when control fields designed for robustness will be self-similar to those designed without robustness considerations?

It is suggestive that self-similar robust fields will exist for weak disturbances, but there is presently no proof of this conjecture.

The results of questions 19–23 may form part of the answer to the central question:

Question 24. Can a general analysis be performed on the sensitivity of the cost functional to variations in the field free Hamiltonian, coupling operator, and control field to determine in what cases to expect significant robustness to environmental fluctuations?

For example, the potential and dipole operators critically determine the control law, and the answers to question 24 should provide information into how errors in these functions influence the control quality.

The robustness of *coherences* for controlled quantum systems in mixed states (i.e., the robustness to decay of the off-diagonal terms in the density operator) is a topic of special interest. The effects associated with this decay are especially important in the quantum information sciences [87], where the development of methods to curtail decoherence in information processing algorithms is an active area of research [88,89]. A relevant contribution would be the combination of optimal control techniques with the ideas of decoherence-free subspaces [90–92], in which dynamics are invariant to interference that would otherwise cause coherences to decay (this is related to the general notion of disturbance decoupling in mathematical systems theory, cf. [22]. The result would be control solutions that maximize coherences while simultaneously minimizing an objective cost functional. While in some cases decoherence free subspaces may not rigorously exist [93], these considerations suggest seeking a general formulation:

Question 25. Can a min–max optimal control problem be posed which maximizes coherence (i.e., maintains the off-diagonal terms in the density matrix) while optimizing the objective functional in such a way that solutions are restricted to lie in a particular subspace? Can such a subspace be identified that is valid in a particular system for a general class of objective operators?

The optimal coherence-control subspaces identified by an analysis following question 25 could be of significance in developing a physical understanding of the mechanisms of decoherence and its suppression. In particular, for systems with well-defined decoherence free subspaces identified from the Hamiltonian, an interesting comparison could be made with the optimal coherence-control subspaces. Other schemes are also being considered for the dynamic manipulation of decoherence and control in the presence of dissipation [94–97].

For applications in which the persistence of coherence is not so evidently important, a complementary line of investigation may seek to determine the general relevance of coherence in accomplishing control objectives. In this regard a case of special interest is the control of *condensed* phases. In an n-dimensional space $\mathcal{H}_n$ and in the presence of rapid dephasing (which implies vanishing of the off-diagonal coherence terms of the density matrix), equation (6) reduces to a set of rate equations for the population in the n states. Under certain conditions, successful controls can be designed for quantum systems whose evolution is determined by these rate equations [98]. Thus, it is evident that full coherence may not be operative in the control mechanism for some systems, and this raises a basic question:

Question 26. Can quantitative measures of coherence be developed to assess its role in any quantum control problem?

The answer to this question will arise from seeking observables that are sensitive to off-diagonal elements of the density matrix.

The application of control methods to the cooling of quantum systems is an active area of research [38–41], and the final question of this section seeks to address the relevant effects of uncertainties in the associated control law. There are several means of defining cooling on the molecular scale. One typically utilized criterion [39] aims to minimize the von Neumann entropy $\sigma = -\sum_k p_k \log p_k$ corresponding to some observable O (such as the system Hamiltonian); here, p_k is the probability that the system is in the kth eigenstate of O. Another system cooling criterion is to *increase* in the Reyni entropy $\mathrm{Tr}(\rho^2)$ [39]. With both measures, maximal cooling is achieved when all but one of the p_k are zero (i.e., achievement of a pure state). Thus, the ability to completely cool a molecular system is likely to be a challenging task in the presence of laser noise. For the purposes of molecular cooling, a laser control with noise fluctuations may be thought of as having an effective nonzero "temperature". Thus a basic question arises about the degree of molecular cooling possible by means of a noise contaminated control field:

Question 27. Can lower bounds be established beyond which a laser characterized by nonzero noise fluctuations cannot cool a quantum system?

2.2.3. Tracking theory

There generally exist a multiplicity of solutions to the quantum optimal control equations (cf. question 14), suggesting that it may be possible to *predefine* a selected path between the initial and final conditions satisfying the control objectives (1). The existence of such a path exactly matching the conditions at both ends assumes that the system is controllable. The path can be implicitly defined by the expectation values $y(t)$ of a tracking operator O_{tr}:

$$y(t) = \langle \psi(t) | O_{\mathrm{tr}} | \psi(t) \rangle, \quad t \in [0, T]. \tag{18}$$

The quantum tracking control problem [99–102] may be viewed as a special case of optimal control theory with the target being the expectation value of O_{tr} over the entire time interval. (In some cases it may be physically attractive to only require that $\lim_{t \to T} O_{\mathrm{tr}}(t) = O$, where O is the objective operator whose expectation value is desired at T.) Given the path defined in equation (18), the tracking algorithm for determining the control law may be derived from the Heisenberg equation of motion

$$\mathrm{i}\hbar \frac{\mathrm{d}\langle \psi(t) | O_{\mathrm{tr}} | \psi(t) \rangle}{\mathrm{d}t} = \langle \psi(t) | [H, O_{\mathrm{tr}}] | \psi(t) \rangle + \langle \psi(t) | \frac{\partial O_{\mathrm{tr}}}{\partial t} | \psi(t) \rangle. \tag{19}$$

With a control law of the form $C(t) = -\mu\varepsilon(t)$ and the assumptions that O_{tr} is independent of time along with $[\mu, O_{\mathrm{tr}}] \neq 0$, equation (19) can be rewritten to solve for the electric field:

$$\varepsilon(t) = \frac{\mathrm{i}\hbar\, \mathrm{d}y/\mathrm{d}t - \langle \psi(t) | [H_0, O_{\mathrm{tr}}] | \psi(t) \rangle}{\langle \psi(t) | [\mu, O_{\mathrm{tr}}] | \psi(t) \rangle}. \tag{20}$$

This equation may be substituted into the Schrödinger equation (3), which then can be numerically solved for $\psi(t)$; substituting $\psi(t)$ back into equation (20) gives an explicit expression for the required control law. One important feature of this technique is that it requires only a single numerical solution of the Schrödinger equation, as opposed to the iterative methods of standard optimal control.

Given freedom in the selection of $y(t)$, one might unknowingly choose a track that generates one or more singularities, or events at which the denominator of the control field in equation (20) vanishes. This type of singularity may be classified as trivial [103] if it exists for all $t \in [0, T]$. Trivial singularities may be removed by formulating a tracking equation analogous to equation (20) for control of the kth time-derivatives of $y(t)$. A rank index may be assigned to each tracking singularity by determining the smallest order k_r for which the corresponding tracking equation has no trivial singularity; if the rank index is infinite, then the track–system pair is uncontrollable. Otherwise, any remaining (isolated) singularities may be treated as nontrivial singularities of some relative order k_{nt} [103]. The magnitude of the disturbance to the trajectory resulting from a nontrivial singularity depends inversely on the magnitude of the derivatives $\partial^i y/\partial t^i$ for $i \leqslant k_{\mathrm{nt}}$ evaluated at the singularity. This partially explains the effects of singularities on quantum tracking control, and invites the question:

Question 28. Can a non-iterative algorithm be developed to sense the occurrence of a forthcoming singularity and accordingly alter the path to avoid the momentary singularity while eventually reaching the objective?

Several extensions of exact inverse tracking which relax demands that could otherwise produce physically unreasonable fields have been developed [104]. The first of these methods is local track generation, in which the problems associated with an *a priori* trajectory design are avoided by letting the track depend on the evolving quantum state: $y(t) = y(\psi(t))$. This approach is especially useful when the control objectives are not specifically defined by target operator expectation values as in equation (1), but rather can be expressed as the production of some qualitative change in a system. A second method is asymptotic tracking, in which the operator O_{tr} is modified to allow an asymptotic approach to possibly singular trajectories. Finally, in the competitive tracking technique a cost functional is defined whose minimization produces a solution optimally matching a *number* of trajectories for different tracking operators as well as minimizing the field fluence or satisfying other control objectives. There is considerable room for further development of the tracking procedure guided by the attraction of performing only one solution of the Schrödinger equation to achieve a control design. Moreover, thus far tracking control has only been applied to the wave function formulation of quantum mechanics. A significant extension would be to treat mixed states:

Question 29. Can quantum tracking control be extended to the density matrix formulation?

In this context, the expectation value $\langle O_{\rm tr}(t)\rangle = {\rm Tr}(\rho(t)O_{\rm tr})$ would be followed and the Schrödinger equation would be replaced by equation (6), with the possibility of additionally including decoherence processes.

2.2.4. Classical mechanics formulations

Up to this point almost all of the questions in this paper have focused on *quantum* dynamics descriptions of atomic and molecular systems. Classical modeling of quantum systems is a common and often successful technique, and it should have a level of applicability in molecular control. The next set of questions attempts to assess this applicability, or the quantum character of molecular scale control. Nonclassical characteristics of dynamical behavior include tunneling, quantization of energy levels, and interference processes, suggesting the question:

Question 30. What characterizations of nonclassical behavior are relevant to defining the quantum nature of a control problem? Can some of these measures be used to estimate the loss of reliability (i.e., defined upon comparison to the analogous quantum system response to the classically designed field) in resorting to the classical optimal control formulation?

Some aspects of this question have been addressed [105], where quantum $C_{\rm q}(t)$ and classical $C_{\rm c}(t)$ control laws corresponding to equivalent representations of specific control problems are compared. The equations of motion analogous to equation (3) are:

$$\frac{{\rm d}q_i^l}{{\rm d}t} = \frac{\partial H}{\partial p_i^l}, \qquad \frac{{\rm d}p_i^l}{{\rm d}t} = -\frac{\partial H}{\partial q_i^l}, \tag{21}$$

and expectation values for the classical system are given by

$$\langle O_{\rm c}\rangle = \sum_{l=1}^{N_{\rm c}} \Gamma_l \overline{O}(\mathbf{q}^l, \mathbf{p}^l), \tag{22}$$

where i ranges over the particle coordinates, l indexes initial conditions $(\mathbf{q}^l(0), \mathbf{p}^l(0))$, and the weights Γ_l for the $N_{\rm c}$ initial conditions are chosen to mimic as best as possible the probability distribution function for the corresponding quantum system. Here, $\overline{O}$ is a classical observable corresponding to its quantum analog. It should be noted that the ordinary differential equations in (21) for some cases may be more expensive to solve than their quantum counterpart in equation (3). One motivation for considering classical optimal control design is for the physical insight possible with classical mechanics.

Optimal control theory has been used [105] to separately design a control field $\varepsilon(t)$ that minimizes the difference between $\langle O\rangle$ and $\langle O_{\rm c}\rangle$ *and* the difference between each of these expectation values and the control objectives on $\mathcal{T}$. For the example of a Morse oscillator, it was found that an optimal control law designed in this fashion produced very close agreement between $\langle O\rangle$ and $\langle O_{\rm c}\rangle$. This result suggests that in some cases classically-designed controls can also be successful as quantum controls. In related

work [106], a method was developed for determining potentials under which evolving classical and corresponding quantum systems give similar values of classical and quantum observables; the approach met with considerable success for the control of dissociative flux and displacement. These developments inspire the question:

Question 31. For what general classes of Hamiltonians and control objectives can the quantum control problem be adequately addressed using the classical equations of motion?

Because interference itself is a nonclassical phenomenon, questions 30 and 31 are related to the considerations of decoherence in the previous section.

3. Laboratory achievement of closed loop control

The design of control laws poses interesting theoretical challenges, and the practical motivation for such a task is to accomplish successful control in the laboratory. In this spirit, this section discusses the conceptual and theoretical aspects of laboratory operations in which information about the evolving quantum systems is used to improve or define effective control laws. Section 3.1 will cover the technique of quantum learning control, which is increasingly proving to be the most efficient method of practically achieving many control objectives, especially in complex quantum systems. Sections 3.2 and 3.3 discuss aspects of feedback quantum control. Learning and feedback control are closed loop experimental procedures aimed at achieving control even in the presence of Hamiltonian uncertainties and laboratory disturbances.

3.1. Quantum learning control

The computational design of a control law to meet a physical objective requires (i) explicit knowledge of the system Hamiltonian and (ii) the ability to numerically solve the quantum control equations at least once (in the case of tracking control) or many times for convergence to an optimal solution. In practice, however, these requirements can rarely be met. If the system to be controlled is sufficiently complex (e.g., a polyatomic molecule), it is likely that the Hamiltonian will be only approximately known and the corresponding quantum design equations can only be solved under serious approximations. In light of these limitations, a completely different and practical approach to the control of quantum dynamics phenomena was developed [107]. In this quantum learning control technique, the laboratory quantum system in itself serves as an analog computer to guide its own control. This approach addresses the requirements of (i) and (ii) above: a physical quantum system can solve its Schrödinger equation in real time and with exact knowledge of its own Hamiltonian, all without computational cost to the user. Hence, the burden of knowing the Hamiltonian and solving the Schrödinger equation is shifted over to a laboratory effort with a learning algorithm guiding the control experiments. The number of physical/chemical systems treated in this way is growing rapidly,

and in many cases it is easier to do the experiments than perform the designs. However, this approach can still benefit from even approximate control designs to start the laboratory learning process, and theory also has an important role to play in introducing the proper stable and reliable algorithms to make the experiments successful.

In summarizing the methodology of quantum learning control, we will take the set of objective times in (1) as discrete and finite: $\mathcal{T} = \{t_i\colon\ i = 1, \ldots, n\}$. The first step is to prepare the laboratory quantum system in a convenient initial state $\psi(0) = \psi_0$ or distribution of incoherent states specified by $\rho(0)$. Next, the system is allowed to evolve under its Hamiltonian and some initial trial control law $C_0^1(t\colon\ t \in [0, t_1])$ applied in the laboratory. At the time t_1, a measurement of the corresponding control objective(s) is made. The quantum system (perturbed by this measurement) is then discarded, and the control law may be updated to $C_1^1(t\colon\ t \in [0, t_1])$ based on the information gained through this measurement. The method and the frequency with which the control law is updated depends on the specific learning algorithm being used (e.g., as described below, with a genetic algorithm the control law is updated after some multiple of N_{pop} experiments in each time interval).

This updating continues until the learning algorithm has converged to some final control law $C^1(t\colon\ t \in [0, t_1])$. At this point, a new initial control law $C_0^2(t\colon\ t \in [t_1, t_2])$ is defined on the next time interval, and an identically-prepared quantum state ($\psi(0) = \psi_0$) is allowed to evolve under $C^1(t\colon\ t \in [0, t_1]) \cup C_0^2(t\colon\ t \in [t_1, t_2])$. The procedure is repeated for the remaining $n-1$ timesteps. In general, physically realizable control laws must be continuous in time, so the C^i are constrained to match on their boundaries. At the conclusion of the process, a control law $C(t\colon\ t \in [0, t_n]) = \bigcup_{i=1}^{n} C^i(t\colon\ t \in [t_i, t_{i-1}])$ will be determined that achieves the objectives (1) within the convergence bounds of the learning algorithm.

The methods most widely used to accomplish the updating of learning control laws are genetic algorithms (GAs) [108], although other learning algorithms could be used. A GA involves the evolution of successive generations of control laws from their parents, in some fashion mimicking biological evolution. Each trial control field is digitized to form a gene which is part of an overall population subject to evolution in the laboratory in search of an optimal control to meet the proposed objectives. Specifically, experiments are performed in which the quantum system evolves under each of the N_{pop} members of a control law "population" $\{{}^lC_j^k(t\colon\ t \in [t_{k-1}, t_k]); l = 1, \ldots, N_{\text{pop}}\}$, where j is the generation number and k indicates the sequential time interval on which the population is defined. The "fitness" of the control law ${}^lC_j^k(t\colon\ t \in [t_{k-1}, t_k])$ is evaluated based on the degree to which the control objective(s) are achieved, and the fittest control laws are preserved, crossed over, or randomly mutated in some prescribed fashion in the next generation. The cross over operation entails recombining complementary sections of the control laws. This procedure is continued until the fittest members of the control law population achieve the control objectives to the required extent. Once this is achieved, the index k is increased and the method is repeated for the next time interval (although all the experiments carried out thus far, e.g., [1,2,4–6,109,110], have been done with a single time interval $t \in [0, t_1]$, $t_1 = T$).

The power of the GA lies in its ability to globally search the space of control laws and discover solutions that possibly may be highly nonintuitive. This directed search takes advantage of the ability to perform a great number of distinct control experiments in a short period of laboratory time, and the closed loop technique has been demonstrated for a wide variety of quantum systems and control objectives. The method has also been shown to have surprising convergence properties. For example, it is observed in simulations [107] and the experiments, e.g., [1,2,4–6], that the GA algorithm can converge for a set of *randomly* constructed initial control law populations. These results suggest the question:

Question 32. Can the necessary physical criteria be identified in order for learning control algorithms to rapidly converge from arbitrary initial control laws? To what extent will the convergence properties of these algorithms be improved by incorporating trial designs or other physical information into the initial fields?

The choice of cost functionals used in the experiments has the same freedom as for computational optimal control theory except that in laboratory learning control there is no direct access to the wave functions. At present the experiments have considered only the final target in the cost, but other criteria could be included giving rise to a possible enhancement to the procedure:

Question 33. Considering that the quantum optimal control problem generally has many solutions, what are the effects of incorporating more extensive fitness criteria (besides the target) into genetic algorithms to select against undesirable aspects of control fields or other observed dynamical features?

Simulations have considered the effects of laboratory errors (modeled as distortions, or transformations, of true input and output data) and noise upon the learning control process [111,112]. In [112], input errors were modeled by performing various functional transformations on the control laws ${}^{l}C_j^k(t\colon\ t \in [t_{k-1}, t_k])$ used in simulated experiments, while output errors were represented by transforming the expectation values of the control objectives corresponding to these experiments. In general, if the input errors are systematic and the output errors are random, they may not significantly affect the ability of the learning algorithm to find an optimal solution. The fitness of the final control laws found by the GA are also demonstrated to be reasonably insensitive to noise in control fields. These results are based on very limited studies of simple model systems, and they invite further investigation:

Question 34. Can stability analyses be performed on closed loop quantum learning algorithms under uncertainties and disturbances in the measurements and control fields for general classes of quantum systems and control objectives?

Such an analysis could give insight into how best to operate the laboratory experiments.

In principle, any optimization algorithm can be applied to quantum learning control. For example, gradient descent and simulated annealing algorithms have been explored in simulations [111], but the GA outperformed them in several test cases. However, this subject has not received a thorough examination:

Question 35. Do there exist algorithms that converge with greater efficiency or robustness than the genetic algorithm for certain classes of quantum mechanical learning control problems?

In treating question 35 it is important to consider the ability to perform very large numbers of quantum control experiments, which may overcome certain algorithmic shortcomings found under more common conditions. This ability is almost unprecedented in other applications of learning algorithms.

Another approach to quantum learning control is provided by the use of input $\rightarrow$ output mapping techniques [113,114]. These methods develop an effective map between the inputs (i.e., the parameters or features defining the control laws) and the outputs (i.e., the expectation values of objective operators). A map from the control input space $\mathcal{C}$ to the space of possible expectation values may be determined directly from the laboratory input and output data; a series of these maps may be needed to cover a sufficiently large portion of $\mathcal{C}$. The control law that optimally satisfies the objectives can be identified from these maps using a suitable learning algorithm. Logical next steps in the development of these methods include answering:

Question 36. What methods can be used to extend the linear input–output learning control techniques developed in [113,114] to generate nonlinear maps?

Question 37. Can a robustness analysis of maps combined with learning control algorithms be performed to study the convergence of these algorithms in the presence of noise and measurement uncertainty?

Progress toward answering question 36 has recently been made in [117]. In considering questions 36 and 37 a central issue is establishing the efficiency of mapping techniques as compared with eliminating the maps altogether in favor of having the learning algorithm directly interfaced with the laboratory experiments. Beyond issues of efficiency, mapping techniques may offer the additional benefit of providing physical insight into control mechanisms based on the observed map structure.

3.2. Feedback quantum control I: The feedback nonlinear Schrödinger equation in the absence of measurement effects

This section concerns the behavior, stability, and classes of solutions to the (deterministic) continuous-feedback Schrödinger equation, where the algorithm operates in

a fashion such that observations do not disturb the control process (i.e., the absence of measurement effects). In the limit that the interval between successive measurement times $\{t_1, t_{2,\ldots}\} \equiv \mathcal{T}_c$ tends to zero and other suitable conditions, the continuous-feedback Schrödinger equation conceptually follows from an infinite sequence of laboratory experiments. In this approach, the probabilistic effect introduced by a measurement at a time t_i is avoided by "discarding" the quantum system after it has been allowed to evolve on the interval $[0, t_i]$ under the control law $C(t) = \bigcup_{k \leqslant i} C^k(t)$, where the domain of each C^j is $[t_{j-1}, t_j]$. After the measurement is performed at t_i, a new quantum system is prepared in the state $\psi_0 = \psi(0)$, and the process is repeated on the interval $[0, t_{i+1}]$. Since $\psi(t)$ is not itself observable, a physically plausible feedback control law based on the measurements taken at t_i depends on $\psi(t)$ through the expectation values of some observable O_c:

$$C\big(\langle\psi(t)|O_c|\psi(t)\rangle\big) = C\big(\psi(t_i)\big), \quad t_i \in \mathcal{T}_c. \tag{23}$$

In practice, an approximation to the limit of infinitely many measurements discussed above could be attained with sufficiently-small spacing between the t_i and the use of interpolation to calculate the continuous function $\langle O_c(t)\rangle$. In general, the procedure is a special type of learning control where the control law is made to explicitly depend on the observation.

The evolution of the continuous-feedback quantum system is described by the equation

$$i\hbar\frac{\partial\psi(t)}{\partial t} = \big[H_0 + C\big(\langle\psi(t)|O_c|\psi(t)\rangle\big)\big]\psi(t). \tag{24}$$

Different choices of $C(\cdot)$ and O_c may result in equation (24) having qualitatively diverse behavior. An interesting case exists [77] under the assumptions (i) that $O_c = \delta(\mathbf{x} - \mathbf{x}')$ is the Dirac delta operator, and (ii) that the control law is $C(|\psi(\mathbf{x}, t)|^2) = -\gamma|\psi(\mathbf{x}, t)|^2$, where γ is a positive constant. With $H_0 = -(\hbar^2/2m)\nabla^2$, the resulting equation

$$i\hbar\frac{\partial\psi(\mathbf{x}, t)}{\partial t} = -\left[\frac{\hbar^2}{2m}\nabla^2 + \gamma\big|\psi(\mathbf{x}, t)\big|^2\right]\psi(\mathbf{x}, t) \tag{25}$$

admits dispersion-free solutions (i.e., it preserves $|\psi(\mathbf{x} - \mathbf{v}t)|^2$) and also solitonic solutions under suitable conditions [75,76]. These types of stable solutions may be significant in many applications of quantum control, including quantum information theory. Equation (25) may also be derived from the quantum optimal control formalism under the assumptions stated above; thus, dispersion free control solutions are optimal under these same conditions.

The possibility of dispersion free dynamics with special control Hamiltonians does not violate the inherently dispersive nature of quantum mechanics as normally encountered: when closed loop control is present, the Schrödinger equation becomes nonlinear and it becomes possible to manipulate dispersion. In descriptive terms one can think of the control actively "batting about" the evolving wave packet to keep it together as it evolves. The existence of such a control law inspires the question:

Question 38. What general classes of control Hamiltonians $H_0 + C(\langle O_c(t)\rangle)$ can be found that exhibit nondispersive or other distinct types of qualitative behavior of practical interest?

Except for tracking control in equation (20), very little work has been done on control laws of the general form in equation (24). Valuable insight might be gained from such studies as well as the possibility of identifying particularly interesting or effective control laws.

3.3. Feedback quantum control II: The feedback nonlinear Schrödinger equation in the presence of measurement effects

This section is concerned with the effects of taking real-time measurements on a single quantum system *while* it is being controlled over the interval $[0, T]$ (here, "single" implies that the sequential "measure and discard" approach of the previous section is abandoned). This scenario naturally arises in the implementation of a feedback control law where measurements are taken on a discrete set of times $\mathcal{T}_M = \{t_i\}$ and M is an observable operator: the control law may be written as $C(\langle\psi(t_i)|M|\psi(t_i)\rangle) = \mathrm{C}(\psi(t_i))$, $t_i \in \mathcal{T}_M$. Feedback may also augment learning or optimal control methods by providing real-time information about the evolving quantum system for the stabilization of particularly sensitive objectives (e.g., locking a quantum system around an unstable point on its potential energy surface). There exist well-established procedures for determining feedback control laws based on measurements of evolving deterministic and stochastic classical systems in engineering control (e.g., [22,116]) and it is possible that many of these methods may be adapted to quantum mechanical control problems.

Extensive consideration has been given to the effects of measurements on evolving quantum mechanical systems, including analysis in the contexts of continuous feedback and the control of quantum systems by homodyne detection (i.e., measurement of a component of the light field) [117–126]. These works generally treat the more difficult problem of random measurement times; here, we give only an elementary discussion of ideas relevant to feedback control with measurements taken at a determinisitic, discrete set of times $\mathcal{T}_M = \{t_i\}$ and suggest some of the most basic accompanying questions.

A postulate of quantum mechanics states that a *perfectly precise* measurement of an operator M must both yield one of the eigenvalues of the operator and result in a disturbance such that ψ collapses to lie within the associated eigenspace of the operator. If the spectrum of M is degenerate we replace M by a complete set of commuting operators (CSCO) [127] in what follows so that each possible measurement specifies a unique eigenstate of the quantum system. The measurement process introduces a stochastic element into the evolution of the quantum system, with the *probability* that a particular eigenvalue of M is observed depending on the state of the system at the time of the measurement. The analysis below assumes that ψ evolves in an n-dimensional space $\mathcal{H}_n$.

The quantum trajectory description of the evolving system expressed here is a special case of more general treatments [118]. The quantum system is initially taken to be in the pure state

$$\psi(0) = \sum_{k=1}^{n} c_k(0)\psi_k, \tag{26}$$

where the c_k are time-dependent coefficients and the $\{\psi_k\}$ are the eigenstates of M. In addition, we assume that in the absence of measurement effects the system evolves under the Hamiltonian $H(t) = H_0(t) + C(t)$, and that all measurements are perfectly precise. The probability that such a measurement at time t_i will yield the eigenvalue λ_k is denoted by $\mathrm{Prob}\{\lambda(t_i) = \lambda_k\} \equiv \mathrm{P}_{t_i}(\lambda_k)$ and the (equivalent) probability that the state is ψ_k immediately after the measurement at t_i is denoted by $\mathrm{Prob}\{\psi(t_i) = \psi_k\} \equiv \mathrm{P}_{t_i}(\psi_k)$.

With these definitions, we have

$$\mathrm{P}_{t_1}(\lambda_k) = \mathrm{P}_{t_1}(\psi_k) = \left|c_k(t_1)\right|^2, \tag{27}$$

where the $c_k(t_1)$ are the coefficients of the state function at the instant of the first measurement. These coefficients are defined by

$$\sum_{k=1}^{n} c_k(t_1)|\psi_k\rangle = \mathsf{T}\exp\left[\frac{1}{\mathrm{i}\hbar}\int_0^{t_1} \mathrm{d}t'\left(H_0\big(t'\big) + C\big(t'\big)\right)\right]\big|\psi(0)\big\rangle, \tag{28}$$

where T is the time ordering operator. Following [118], we let $|\psi_{\mathrm{c}}(t_i)\rangle$ be the (pure) quantum state *conditioned* on perfect knowledge of the measurement at time t_i; for instance, the state $|\psi_{\mathrm{c}}(t_1)\rangle$ is a random variable with distribution given by equation (27). These definitions may be extended for the remainder of the t_i:

$$\mathrm{P}_{t_{i+1}}(\lambda_k) = \mathrm{P}_{t_{i+1}}(\psi_k) = \left|c_k(t_{i+1})\right|^2, \tag{29}$$

where $c_k(t_{i+1})$ are random variables defined by

$$\sum_{k=1}^{n} c_k(t_{i+1})|\psi_k\rangle = \mathsf{T}\exp\left[-\frac{1}{\mathrm{i}\hbar}\int_{t_i}^{t_{i+1}} \mathrm{d}t'\left(H_0\big(t'\big) + C\big(t'\big)\right)\right]\big|\psi_c(t_i)\big\rangle = \big|\psi(t_{i+1})\big\rangle. \tag{30}$$

Here, there is no subscript c in the last equality, because $|\psi(t_{i+1})\rangle$ represents the state of the system at the instant before the measurement at t_{i+1}.

Introducing the projection operators

$$F_k(t_{i+1}) = |\psi_k\rangle\big\langle\psi(t_{i+1})\big|, \tag{31}$$

the evolution of the particle under the measurements may be written as a "stochastic quantum map" [118] between states at the t_{i+1}:

$$\big|\psi_{\mathrm{c}}(t_{i+1})\big\rangle = F(t_{i+1})\mathsf{T}\exp\left[-\frac{1}{\mathrm{i}\hbar}\int_{t_i}^{t_{i+1}} \mathrm{d}t'\left(H_0\big(t'\big) + C\big(t'\big)\right)\right]\big|\psi_{\mathrm{c}}(t_i)\big\rangle, \tag{32}$$

where the probability $\mathrm{Prob}\{F(t_{i+1}) = F_k(t_{i+1})\} \equiv \mathrm{P}_{t_{i+1}}(F_k(t_{i+1}))$ that the projection at time t_i is $F_k(t_i)$ (i.e., onto the kth eigenstate) is given by equation (29):

$$\mathrm{P}_{t_{i+1}}\big(F_k(t_{i+1})\big) = \mathrm{P}_{t_{i+1}}(\lambda_k) = \mathrm{P}_{t_{i+1}}(\psi_k). \tag{33}$$

We note that equation (32) has both deterministic $H_0(t')$ and random $F(t_{i+1})$, $C(t')$ terms (where the latter are assumed to be a function of the random state $\psi(t)$).

The state of the stochastic system at time t is then described by the density matrix (here, simply a sum of pure states)

$$\rho(t) = \sum_{l=1}^{N_\mathrm{p}(t)} p_l \big|\psi_l(t)\big\rangle\big\langle\psi_l(t)\big|, \tag{34}$$

where p_l is the probability that the lth possible trajectory $\psi_l(t)$ was realized on a given solution of equation (32). Here, $N_\mathrm{p}(t)$ is the number of these possible trajectories taken up to time t. The number of measurements that has taken place before time t is $m(t) = \mathrm{card}(\{t_i\colon t_i \leqslant t\})$, and each combination of these measurements generally defines a unique trajectory so that $N_\mathrm{p}(t) = m(t)^n$. In general, the p_l and, hence, the density matrix given in equation (34) could be approximated by a Monte Carlo simulation of equation (32): if N_MC states $|\psi^a(t)\rangle$ were produced in an ensemble of simulations, then

$$\rho(t) \approx \frac{1}{N_\mathrm{MC}} \sum_{a=1}^{N_\mathrm{MC}} \big|\psi^a(t)\big\rangle\big\langle\psi^a(t)\big|, \tag{35}$$

where the expected accuracy of the approximation increases with N_MC at a rate depending on the p_l and $|\psi_l(t)\rangle\langle\psi_l(t)|$.

In the special case that $C(\psi(t)\colon\ t \in [t_i, t_{i+1}])$ depends only upon t_i and the value $\langle M(t_i)\rangle$ of the most recent measurement, equation (32) defines a Markov chain [128] with (generally) nonstationary transition probabilities. For any t_i and each k, all paths to $|\psi(t_i)\rangle = |\psi_k\rangle$ then lead to the same evolution on (t_i, t_{i+1}), so that summing over $N_\mathrm{p}(t) = m(t)^n$ trajectories is redundant and we may instead set $N_\mathrm{p}(t) = n$. The p_l may, therefore, be calculated from $m \times n^2$ transition probabilities between the states $|\psi_k\rangle$. With the additional assumptions that the t_i are evenly spaced and that the control law is time-independent, the Markov chain becomes stationary with n^2 transition probabilities generating the evolution of the system.

If the measurement process does not completely determine the quantum states (or if the system is initially in a mixed state), the measurement-conditioned system may no longer be described as a sum of pure states as in equation (34). This could occur, for example, (i) due to imprecision in the measurement process resulting from equipment limitations or environmental fluctuations, or (ii) if M were not a CSCO, in which case projection after measurement would be into a (degenerate) space spanned by multiple eigenstates. In such cases, an alternative formulation to equation (30) involving a stochastic map between conditioned density operators $\rho_\mathrm{c}(t_i)$ is required [118].

Each measurement in feedback quantum control involves an information tradeoff: the system is perturbed away from the deterministic Schrödinger equation, but a measurement is used to update the control law. This raises a question similar to that suggested in [11]:

Question 39. In what general classes of problems does incorporating feedback measurements assist the achievement of quantum control objectives? In what cases does it hinder the achievement of these objectives? More specifically, can it be shown for general classes of problems that taking a certain number of measurements improves control (with reasonable assumptions including the final time T being sufficiently large)? Does there exist a (problem-dependent) frequency and timing of measurement to optimize the feedback quantum control problem?

Another question relating to this issue of information tradeoff is:

Question 40. What kind of weak observations give useful information about a quantum system while introducing only minimal perturbations?

As a physical example of a weak observation, consider measurement of a few atoms of a BEC cloud, which introduces minimal disturbance while characterizing a certain measure of the entire system [10]. The nature of weak observations becomes more subtle in the limit of the system consisting of a single atom or molecule. A related question is:

Question 41. What are the effects of measurements on the feedback control process for systems satisfactorily described semiclassically?

This question opens up consideration of possible gray scale behavior ranging from the classical limit where a measurement can be made with vanishingly small impact on the system out through semiclassical behavior and then to the full quantum limit. If some of the issues in question 39 are severely restrictive for quantum feedback control, then it is suggestive that in the semiclassical limit (which generally describes intramolecular motion well) a window of feedback events may exist before the continuing observational disturbances cause loss of control.

Returning to the most basic issue of control, it would also be interesting to investigate:

Question 42. Under what conditions can controllability be proved for feedback-controlled quantum systems?

3.4. Closing the control loop in laboratory hardware through machine feedback

Recent work in acoustics illustrates the possibility of focusing reflected waves back upon their sources [129,130] in an iterative fashion in order to enhance the intensity in

the focal volume. In this work, an array of speaker/microphone units surrounding the target sends amplified, time-reversed acoustic wave packets recorded as reflections of a previous pulse; the time reversal process ensures that these waves focus on the point from which the previous pulse was emitted with a degree of enhancement on each iterative cycle. An analogy of this technique relevant to quantum mechanics might be "reflection" through special measurement devices (e.g., ideally capable of full three dimensional detection from the controlled sample) that could then send modified electromagnetic waves precisely back to an emitting quantum mechanical source to better achieve the control objectives. While the length scale of electromagnetic waves is too large to directly produce a focusing effect analogous to that in time-reversed acoustics, they could function as an intermediary that excites a desired quantum interference pattern which actually accomplishes the focusing at the atomic/molecular scale. The fundamental question invited by this idea is:

Question 43. Can an array of special hardware units be designed that capture electromagnetic waves emanating from the quantum systems and return modified signals to effect an "automatic" achievement of particular control objectives? For what classes of control objectives and quantum systems might such a procedure be possible, and how would the required modification of the electromagnetic signal be determined and created?

This process may be fully quantum mechanical if carried out in a suitable optical cavity, but in general the same closed loop observation/disturbance issues raised in the previous section must be considered here.

Such a machine feedback control mechanism, if achieved, could be characterized as self-controlling. One natural application would be stabilization of an established quantum dynamic state, which would be a type of feedback control as discussed in section 3.3. But, most exciting would be an apparatus that automatically directed the evolution of a system to the desired target. The machine could be viewed as a fully automated laboratory apparatus implemented in a closed loop quantum context, with the hardware actually being part of the system under control. At this juncture such a machine is only a *gedanken* process, but its potential strongly motivates an analysis of the concept.

4. Hamiltonian identification

Knowledge of the Hamiltonian H_0 and the dipole $\boldsymbol{\mu}$ (or other coupling coefficients) is required for control law design and is of fundamental importance to many other applications in chemistry and physics. Section 4.1 concerns dynamical algorithms that invert time-dependent laboratory data to identify these operators. After a discussion of a particular special case, it is noted that the problem of determining $\boldsymbol{\mu}(\mathbf{x})$ or $V(\mathbf{x})$ (where $H_0 = \mathrm{K} + V$ with K being the kinetic energy operator on $\mathcal{H}$) may be generally expressed as an explicit algorithm closely related [100] to tracking control (cf. section 2.2). The

formulation in section 4.1 suggests that the control law producing the underlying quantum dynamics can play an important role in determining the quality of the inverted data. This observation opens up the possibility of developing a closed-loop device that would couple dynamical inversion algorithms with laboratory machinery. Aspects of the development of such an optimal quantum dynamics identification machine are discussed in section 4.2.

4.1. Methods and algorithms for the inversion of quantum dynamics data

There are significant laboratory efforts toward achieving high-resolution diffraction images of evolving molecules [131] and solid state materials. The data from such experiments is the probability density function $|\psi(\mathbf{x}, t)|^2$ (formally, the expectation value of the Dirac delta operators $\delta(\mathbf{x} - \mathbf{x}')$). An algorithm for inverting this type of data to identify $V(\mathbf{x})$ without the expensive requirement of numerically solving the Schrödinger equation has been suggested [132]. The algorithm relies on Ehrenfest's relation:

$$\int_{-\infty}^{\infty} \nabla V(\mathbf{x}) \left|\psi(\mathbf{x}, t)\right|^2 \mathrm{d}\mathbf{x} = -m \frac{\mathrm{d}^2}{\mathrm{d}t^2} \int_{-\infty}^{\infty} \mathbf{x} \left|\psi(\mathbf{x}, t)\right|^2 \mathrm{d}\mathbf{x}, \tag{36}$$

which is an integral equation for $\nabla V(\mathbf{x})$ with the assumption that $|\psi(x, t)|^2$ is available from laboratory measurements. As the equation is linear, it formally does not require iterative methods for its solution. The attractive nature of this integral equation formulation suggests seeking its possible extension to other types of data that may be less directly related to $\psi(t)$:

Question 44. Can non-iterative algorithms be developed for the inversion of data $\langle O_{\mathrm{h}}(t)\rangle$ for observables O_{h} other than the Dirac delta operator?

The key step leading to the algorithm implied by equation (36) was to use $|\psi(x, t)|^2$ to invert $\langle \mathbf{x}\rangle$ rather than working directly with $\langle \delta(\mathbf{x} - \mathbf{x}')\rangle$. Question 44 may not lend itself to such a simple treatment, but this line of thought directly leads to the tracking inversion approach below.

The problem of determining $\boldsymbol{\mu}(\mathbf{x})$ or $V(\mathbf{x})$ may be related ([100]; see also [133] for a different approach) to the problem of determining the control law $C(t)$ that will cause a quantum system to follow a prescribed track (see figure 3). In particular, if the expectation values $y(t) \equiv \langle O_{\mathrm{h}}(t)\rangle$ of a time-independent operator O_{h} are established from a series of observations of an evolving quantum system, the Schrödinger equation (3) and the Heisenberg equation of motion (19) form the pair of coupled (forward–inverse) equations

$$\mathrm{i}\hbar \frac{\mathrm{d}\psi(t)}{\mathrm{d}t} = \left[H_0 - \boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}(t)\right]\psi(t), \tag{37}$$

$$\mathrm{i}\hbar \frac{\mathrm{d}y(t)}{\mathrm{d}t} = \left\langle \psi(t)\right|\left[H_0 - \boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}(t), O_h\right]\left|\psi(t)\right\rangle. \tag{38}$$

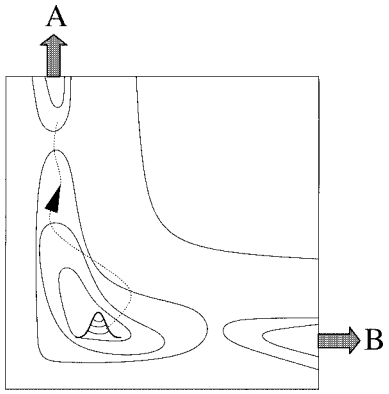

Figure 3. Schematic illustration of wave packet track evolving on a potential surface (contours shown). For control a goal may be to find the field $\varepsilon(t)$ that will steer the track out of product channel A or B, while for inversion the track is observed in the laboratory and the goal is to determine the potential in the regions traveled by the track. From [100], with adapted caption.

The solution of these evolution equations may in principle be attempted for any two unknowns. As knowledge of $\psi(t)$ is not available in any physical problem, the wave function will always be considered as one of these unknowns; the other may be chosen from either $\boldsymbol{\varepsilon}(t)$, $\boldsymbol{\mu}(\mathbf{x})$, or $V(\mathbf{x})$ with the complementary pair assumed as known. The first of these possibilities was treated in section 2.2, where the fact that the expectation value on the right hand side of equation (38) involves only spatial integration was exploited to write $\boldsymbol{\varepsilon}(t)$ explicitly in the (possibly singular) form of equation (20). We now turn to the solution for $\boldsymbol{\mu}(\mathbf{x})$ and $V(\mathbf{x})$.

Considering for simplicity the one-dimensional case, equation (38) may be rewritten [100] as a Fredholm integral equation of the first type:

$$\int_{-\infty}^{\infty} K(x;t)\,f(x)\,\mathrm{d}x = h(t), \tag{39}$$

where if $\mu(x)$ is the unknown variable,

$$f(x) = \mu(x), \tag{40}$$

$$K(x;t) = -\varepsilon(t)\,\mathrm{Im}\big\{\big(O_{\mathrm{h}}\psi(x,t)\big)^{*}\psi(x,t)\big\}, \tag{41}$$

$$h(t) = \frac{\hbar}{2}\frac{\mathrm{d}y(t)}{\mathrm{d}t} - \mathrm{Im}\big\langle\psi(t)\big|\,O_{\mathrm{h}}H_0\big|\psi(t)\big\rangle, \tag{42}$$

and if $V(x)$ is the unknown,

$$f(x) = V(x), \tag{43}$$

$$K(x;t) = \mathrm{Im}\big\{\big(O_{\mathrm{h}}\psi(x,t)\big)^{*}\psi(x,t)\big\}, \tag{44}$$

$$h(t) = \frac{\hbar}{2}\frac{\mathrm{d}y(t)}{\mathrm{d}t} - \mathrm{Im}\big\langle\psi(t)\big|\,O_{\mathrm{h}}\big(\mathrm{K} - \varepsilon(t)\mu(x)\big)\big|\psi(t)\big\rangle. \tag{45}$$

It is apparent that there does not exist an explicit formula for $f(x)$ analogous to that given in equation (20) for $\varepsilon(t)$: this difference lies in the distinct roles of space and time in quantum mechanics. Equation (39) is ill-conditioned, and even for nonvanishing kernels $K(x;t)$, a unique solution may not exist. Thus, equation (39) generally requires regularization before it can be solved for *f(x)*. This may be accomplished, for example, through minimization of the cost functional

$$J = \int_0^T \left[\int_{-\infty}^{\infty} K(x;t) f(x)\,\mathrm{d}x - h(t) \right]^2 \mathrm{d}t + \alpha \int_{-\infty}^{\infty} f^2(x)\,\mathrm{d}x, \tag{46}$$

where α is a regularization parameter [100] and $[0, T]$ is the control interval. Setting the first variation of J with respect to $f(x)$ equal to zero yields

$$\int_{-\infty}^{\infty} \kappa\left(x, x'\right) f\left(x'\right) \mathrm{d}x' + \alpha f(x) = \widetilde{h}(x), \tag{47}$$

which is a (regularized) Fredholm equation of the second type with

$$\kappa\left(x, x'\right) = \int_0^T K(x;t) K\left(x';t\right) \mathrm{d}t, \tag{48}$$

$$\widetilde{h}(x) = \int_0^T K(x;t) h(t)\,\mathrm{d}t. \tag{49}$$

Solution of the regularized pair (37) and (47) may be accomplished using the tracking procedure discussed in section 2.2, with the role of $\varepsilon(t)$ replaced by $\mu(x)$ or $V(x)$. The procedure consists of formally solving equation (47) for $f(x)$ and substituting the result into the Schrödinger equation (37) to solve for $\psi(t)$; the process needs to be iteratively performed due to the nonlinear manner that $f(x)$ and $\psi(t)$ enter. There are many variants on iterative methods for solving these equations and care is needed to assure that the process is stable.

In general, the goal is to solve equations (37) and (38) with minimum of distortion introduced by additional criteria; in the example above, the balance between this objective and stability requirements is set by α, whose optimal value could be set or approximated based on the details of the particular problem and solution method. In this light, it may be useful to consider:

Question 45. Can (assumed) knowledge of $|\psi(x,t)|^2$ be used to guide the regularization process? For example, since the kernel $\kappa(x,x')$ is trivially singular where $|\psi(x,t)|^2 = 0\ \forall t \in [0,T]$, will the quality of inversion improve if the regularization parameter $\alpha(x)$ is made spatially dependent and scaled inversely with $\int_0^T |\psi(x,t)|^2\,\mathrm{d}t$?

This could be accomplished, for example, by setting $\alpha(x) = \left(\int_0^T |\psi(x,t)|^2\,\mathrm{d}t\right)^{-\beta}$ for some $\beta > 0$.

An important feature of the inverse problem of solving equations (37) and (38) for μ or V is that the evolution of the quantum system over $[0, T]$ which deter-

mines $\kappa(x, x')$ is in turn governed by the applied field $\varepsilon(t)$ in equation (37). Hence, it should be possible to determine a control law which allows inversion with maximum stability to produce optimal dynamical regularization. This comment opens up the more general notion of optimally controlled inversion introduced in section 4.2.

4.2. Dynamical regularization and the realization of an optimal dynamics identification machine

As noted in the previous section, because $K(x, x')$ in equation (48) is trivially zero in spatial regions which are not explored by the wave function at some time during the control period $[0, T]$, meaningful inversion of equations (37) and (38) may only be expected if the control law $\varepsilon(t)$ steers the wave function to be nonzero in the domain in which μ or V is to be determined. It is important to note that the formulation of equations (37) and (38) may be extended to incorporate multiple realizations of the control law $\varepsilon_j(t)$ [100], as this process allows for the inclusion of data $y_j(t)$ from multiple experiments that, taken together, may provide the desired evolution over the entire spatial domain of interest.

For dynamical reasons the kernel $\kappa(x, x')$ may still produce a singular operator in equation (47) where it is significantly non-zero. The additional dynamical regularization conditions required to resolve this problem are not immediately apparent, suggesting the question:

Question 46. What general conditions on the control law in equation (37) can be found that maximize the quality of the solution for $f(x)$ in the inverse equations (37) and (38)?

Some simple observations from classical dynamics may provide insight into these dynamical regularization conditions. The evaluation of observable functions of a classical system along a trajectory does not involve spatial integration, so that the inverse problem analogous to equations (37) and (38) for a classical system is free from the accompanying nonuniqueness problems (i.e., one can simply perform the inversion along the desired trajectory). This suggests:

Question 47. Will control laws favorable for optimal dynamical regularization operate by maximally localizing wavepackets?

With these questions in mind, a complete laboratory device may be envisioned to function as an optimal dynamics inversion machine for the efficient and automatic discovery of V or $\boldsymbol{\mu}$ for diverse quantum systems [134,135]. This machine (or possibly a family of these machines) would operate in a closed-loop mode to take advantage of the ability to perform a very large number of high throughput control-observation experiments. The successful development of such a machine could have a significant impact in other areas, as the concept should be transferable beyond quantum mechanics.

The components of the optimal dynamics identification machine concept are sketched in figure 4:

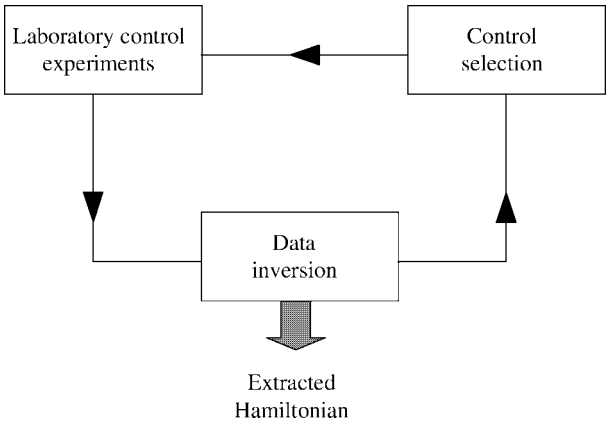

Figure 4. The optimal dynamics identification machine.

(1) Initial approximations for V and $\boldsymbol{\mu}$ could be used to design an optimal control field aimed at causing the wave packet to evolve in desired spatial areas where V and $\boldsymbol{\mu}$ are being sought.

(2) Laboratory experiments using this control law would be performed to produce the data trajectory $\langle O_{\mathrm{h}}(t)\rangle = y(t)$ (various possible ultrafast observations and multiple trajectories could be considered as sources of data).

(3) An inversion would be performed to produce updated potential or dipole information.

(4) If the spatial domain of interest was not completely covered by the current trajectory or if the inversion quality is not adequate, the procedure would be repeated. The partial Hamiltonian information gained from step 3 could assist in the design of an improved experimental control field.

This theoretical embodiment suggests:

Question 48. Can a functional optimal dynamics identification machine be realized which would use designed control fields to incrementally and optimally discover the Hamiltonian for a quantum system?

A positive answer to question 48 would require the resolution of several of the questions posed in the other sections of the paper, as it combines challenges associated with control law design, data inversion, and laboratory implementation of quantum control. Furthermore, to be operational the components of the machine in figure 4 need to work in sync with each other.

We conclude the section on Hamiltonian identification by exploring a *possible* connection between dynamical inversion techniques and the corresponding time-independent eigenvalue problems of traditional spectroscopy:

Question 49. Are new methods for inverting existing time-independent, high-quality spectral data possible which synthesize tracks of the form $y(t)$ and use these tracks in dynamical inversion algorithms?

Tracks of this type can potentially be developed from known Franck–Condon factors and the associated transition energies [100].

5. Identification of quantum control rules of thumb

A cornerstone of chemistry is that physically similar molecules tend to exhibit similar chemical behavior. The emphasis is on "similar", and in the context of quantum control the criteria for defining similarity is not known. From the rich behavior and information content in the design, closed loop, and dynamical inversion aspects of quantum control, one can anticipate using the emerging results to provide insight or estimates for the control laws for physically related, but as yet uninvestigated, problems. The body of relationships (as just yet beginning to be observed) between quantum systems, control objectives, and control laws may be called quantum control rules of thumb. A special example is the explanation of the timing of the pulses used in the STIRAP control method [8]. However, attempts to find general control rules of thumb have proved much more difficult than was at first expected. Part of the difficulty in finding quantum control rules of thumb arises from the existence of multiple solutions to virtually all quantum control problems, especially in the strong field nonlinear regime.

A natural strategy for identifying rules of thumb might ensue from a type of quantum mechanical reverse engineering: solutions $\{C(t), \psi(t), \lambda(t), \langle O\rangle\}$ to the optimal control equations, or $C(t)$ and $\langle O\rangle$ from closed loop experiments, could provide a physical basis for understanding the *mechanisms* and pathways leading the quantum system from initial conditions to final control objectives. However, there exist many examples in the literature in which the structure of the final control fields and the resulting control pathways are found to be highly nonintuitive, and judging the relevance of such solutions in terms of general rules of thumb is difficult in the presence of a possibly large number of (locally) optimal solutions. Further insight into the structure of these local minima may be gained by identifying the family of locally optimal control solutions and enumerating them based on their optimality. This problem might be partially alleviated by incorporating a global search procedure in the optimization algorithm (e.g., a genetic algorithm), both for theoretical design and laboratory control. This practical issue inspires the general question:

Question 50. In light of the existence of multiple solutions to typical quantum control problems, can broadly useful methods be developed to extract information about systematic classes of mechanisms for achieving control?

If such techniques were developed, the existence of multiple solutions could possibly be exploited as a large body of data about control behavior.

In the context of closed-loop laboratory implementation of controls, the presence of multiple solutions to the quantum optimal control problem opens up several options [65]. Given that there exist many possible solutions $C(t)$ from which identification of control mechanisms could be attempted, it is important to select solutions that contain a minimum of extraneous information that detract from this task. In addition, control rules of thumb would best be developed based on solutions that are robust to realistic laboratory noise. Both the suppression of extraneous structural components in $C(t)$ and the selection of robust control fields may be accomplished through the use of appropriate cost functionals [65]. This "cleanup" of control laws is likely to assist in identifying rules of thumb for the control of quantum systems.

A first step toward answering question 50 involves the effective classification of similarities and differences between molecules in a context relevant to the controls directing them to certain physical objectives. This type of classification is fundamental in many fields of chemistry and physics, in which the vast numbers of molecules are categorized according to their relevant behaviors or properties. However, presently the standard measures have not been able to consistently predict the structure of control fields for particular objectives, suggesting the question:

Question 51. For purposes of control, what are the characteristic variables relevant to codifying similarities and differences between atoms or between molecules?

A three-way classification structure will be necessary, relating (i) control laws, (ii) molecular Hamiltonians and coupling terms, and (iii) control objectives. An initial step toward building this structure will likely be identification of the relevant properties of control laws $C(t)$:

Question 52. The identification of spectral components and intensities is a simple way to measure the physical mechanisms encoded in control fields. Considering that the interpretation of these quantities requires knowledge of the underlying Hamiltonian, are there better measures of similarities and differences between control fields?

Insights into control rules of thumb may also be evident from exploring the relation between Hamiltonian structure and control results:

Question 53. Is there a general formulation to estimate how control fields for equivalent objectives will differ under specific classes of Hamiltonian?

One way to answer this question might be through numerical optimal control calculations for a series of quantum systems whose Hamiltonians differ by small increments, but collectively cover a broad sampling of physical systems. Such an experiment would be expensive computationally, although the burden would be eased by exploiting the fact that the controls should vary incerementally from case to case. The results may yield important information relating the structure of optimal control solutions with features of the

corresponding Hamiltonians. Suitable quantum dynamics approximations may also be helpful in achieving this objective.

It would be surprising if answers, at least qualitatively, do not come forth to the posed questions of control mechanism identification and molecular controls classification. Nevertheless, all evidence currently suggests that this will be a very difficult task. The implications of finding quantum rules of thumb for both the theory of quantum control and its practical implementation are very substantial: resolution of this matter may be the most important challenge ahead for the field.

6. Conclusions

The purpose of this paper is to advance the field of control over quantum mechanical behavior by stimulating investigation along a broad set of lines through presentation of a set of questions. As the field evolves some of these questions may prove to have easy answers, while others may eventually be set aside as ill-advised. Other questions and issues may stand as serious hurdles for the development of some aspects of the quantum control field. Regardless of the specific outcome, if this paper serves as a stimulus for the subject, then it will be a success.

Quantum mechanical control is a fast-growing research area, and this natural evolution will likely produce important new questions and challenges. These developments should be welcomed, and in this spirit we encourage the reader to fill in the next question.

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