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The role of theory in the laboratory control of quantum dynamics phenomena

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Abstract. Interest in the control of quantum dynamics phenomena has grown in recent years, with laboratory studies showing increasing successes. The role of theory in the control of quantum phenomena encompasses the design of laser controls, the development of algorithms to guide the laboratory studies, and the means to analyze the ensuing dynamics observations. Laboratory laser control instrumentation has the special capability of performing massive numbers of experiments in a short period of time, to rapidly search for controls that meet the objectives. This unique laboratory feature needs to be factored in when considering how to best utilize theoretical analyses. The present paper reviews the role that theory is playing, as well as suggests some future avenues for theory in the laser control of quantum phenomena.

Keywords: Quantum dynamics – Control – Lasers – Theoretical chemistry

1 Introduction

A longstanding dream has been to steer about dynamical events at the atomic and molecular scale, using the special capabilities of lasers. This dream may be traced back to the earliest days of laser development [1, 2] in the 1960s, and at that time, the features of lasers thought to be significant were their monochromatic character and high focal intensity. The thinking along these lines was only marginally different from that in more traditional photochemistry, except with the hope that the resonantly interacting radiation would induce a particular excitation to selectively break chemical bonds.

The early ventures into laser control over molecular processes largely yielded frustration and a general sense that intramolecular energy transfer was working against successful control. Starting in the late 1980s, it was realized that the key scientific principle underlying control is the active manipulation of constructive and destructive quantum wave interferences [3, 4, 5]. Although certain applications may be amenable to simply utilizing interference along two dynamical pathways [3], the most general way to discriminate amongst complex molecular objectives is through many pathways [4, 5]. Utilizing many pathways allows the highest degree of constructive interference in the desired product state while simultaneously creating destructive interferences in the other undesired states. In addition, operating with strong fields can utilize dynamic power broadening to overcome the need for precisely matching the laser spectrum to the molecular transition frequencies [6, 7]. A number of successful recent experiments operate in this regime [6, 7, 8, 9, 10].

A general paradigm for employing theory to design quantum control fields involves three steps.

1. Obtain the system Hamiltonian and dipole, or other radiative coupling coefficients.
2. Perform appropriate calculations with the Schrödinger equation to design the control laser field taking into account all of the associated physics and other criteria
3. Transfer the design into the laboratory for implementation on the actual sample to be controlled.

Although, in principle, this procedure is viable, it is not the paradigm currently being followed by the increasing number of control experiments [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17], especially for the manipulation of complex quantum systems. The experiments are now being performed largely under closed-loop laboratory learning control [18] that circumvents the theoretical design process. Closed-loop learning control is proving to be generic in its capabilities [18, 19, 20], thereby focusing attention on identifying the future role of theory in this area. This article aims to provide an overview of the field of quantum control, as well as especially to address this latter point. Theory is expected to have an essential and increasing role in the future development of the field; however, this role may take on some unusual characteristics, as it is essential to work with the capability of performing massive numbers of closed-loop experiments

[20]. The present closed-loop experiments grew out of theoretically derived concepts that they be performed in a particular fashion [18, 19], and a number of general considerations suggest that the future developments in this field are likely also to draw on special aspects of theoretical insights and guidance.

The remainder of the paper is organized as follows. An overview of optimal control theory [4, 5, 21, 22] in quantum systems is presented in Sect. 2 as groundwork for the closed-loop learning control experiments [18, 19, 20] described in Sect. 3. Some challenges ahead for theory in the field are laid out in Sect. 4, and concluding remarks are given in Sect. 5.

2 Optimally achieving quantum control objectives

The notion of seeking optimal performance in the exploration of scientific endeavors may at first appear out of place, however, the modus operandi of present day scientific pursuits is permeated with the goal of obtaining the best performance. Examples include the desire for obtaining the highest yield for chemical reactions, the best transition temperature for a superconducting material, the most robust performance of a quantum computing gate, the highest spectral resolution, the best values for rate constants in a chemical reaction, and the best quantitative information about chemical reaction mechanisms. The practice of science is often an effort in optimization, and the boundaries between chemistry, physics, and engineering blur in this regard. Put simply (R. Sauerbrey, private communication), "the best engineering should contain some science and the best science should contain some engineering". Thus, seeking an external field to manipulate a quantum system naturally leads to posing the problem in terms of finding an optimal control field [4, 5, 21, 22]. In the remainder of this paper, these controls will be taken as laser electric fields, although the same concepts apply to other forms of external quantum controls. The physical/chemical phenomena presently considered for control span a wide range, including the manipulation of atomic states [12], molecular bonds for rearrangement [6, 7, 8, 9], semiconductor electron dynamics for switching [11] or other purposes, the performance of gate operations in quantum information systems [23], the selective generation of high harmonics [10], the dynamic discrimination of spectrally similar molecules [17], the creation of specific molecular excitations [14, 24], and the manipulation of biodynamics [15].

The initial formulation [4, 5] (and that treated here, for clarity) of quantum control theory focused on a wave function description that is applicable for systems starting out in pure states and remaining isolated from random environmental interactions [25]. Consider the Schrödinger equation:

$$i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle = [H_0 - \mu\cdot\varepsilon(t)]|\psi(t)\rangle, |\psi(0)\rangle = |\phi\rangle, \tag{1}$$

where H_0 is the field-free Hamiltonian, μ is the dipole moment, and $\varepsilon(t)$ is the laser electric field. A common goal is to seek the field that will take the quantum system from its initial state $|\phi\rangle$ and steer it to yield an optimal value $\tilde{O}$ for $\langle\psi(T)|O|\psi(T)\rangle$ associated with the observable operator O at a specified target time T. Although these statements alone would be sufficient to present a well-posed problem to determine $\varepsilon(t)$, often additional constraints or desires are included. Control may be sought at the lowest value for the laser fluence, $\int_0^T \varepsilon^2(t)\mathrm{d}t$, or minimization of $|\langle\psi(t)|O'|\psi(t)\rangle|$ throughout the time interval $0 \le t \le T$ where O' is an operator associated with a dynamical process whose presence is deemed undesirable (e.g., breaking the wrong bond in control over dissociation). Other criteria may also be introduced regarding the control field, to limit its temporal and/or frequency characteristics in keeping with any laboratory limitations. For the sake of clarity, we will treat a simple case where only the physical objective and laser fluence are considered according to the following design cost function:

$$J' = \left[\langle\psi(T)|O|\psi(T)\rangle - \tilde{O}\right]^2 + \omega\int_0^T \varepsilon^2(t)\mathrm{d}t, \tag{2}$$

where $\omega > 0$ in a weight balancing the two terms. The term $\langle\psi(T)|O|\psi(T)\rangle$is a functional of the sought-after optimal field $\varepsilon(t)$, and the design process may be implemented by considering $\min_{\varepsilon(t)} J'$, subject to satisfaction of the Schrödinger equation in Eq. (1). This constrained optimization problem may be rewritten as an unconstrained one [4, 5] through the introduction of a Lagrange multiplier "state" $|\lambda(t)\rangle$ as follows:

$$J = J' + \int_0^T \Im\langle\lambda(t)|i\hbar\frac{\partial}{\partial t} - H_0 + \mu\cdot\varepsilon(t)|\psi(t)\rangle\mathrm{d}t. \tag{3}$$

The specification of the optimal design problem $\min_{\varepsilon(t)} J$ is now complete using Eq. (3) along with the initial condition $|\psi(0)\rangle = |\phi\rangle$ without any further constraints. A functional variation of Eq. (3) with respect to $\varepsilon(t)$, $|\psi(t)\rangle$and$|\lambda(t)\rangle$ yields the following Euler–Lagrange equations specifying the optimality conditions:

$$i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle = [H_0 - \mu\cdot\varepsilon(t)]|\psi(t)\rangle, |\psi(0)\rangle = |\phi\rangle, \tag{4a}$$

$$i\hbar\frac{\partial}{\partial t}|\lambda(t)\rangle = [H_0 - \mu\cdot\varepsilon(t)]|\lambda(t)\rangle,$$
$$|\lambda(T)\rangle = 2\left[\langle\psi(T)|O|\psi(T)\rangle - \tilde{O}\right]O|\psi(T)\rangle, \tag{4b}$$

$$\varepsilon(t) = \frac{1}{2\omega}\Im\langle\lambda(t)|\mu|\psi(t)\rangle. \tag{4c}$$

These equations have an unusual mathematical structure as a boundary value problem in time through the initial and final conditions in Eq. (4a) and (4b). In addition, they are nonlinear through the final condition in Eq. (4b), as well as the electric field in Eq. (4c). Satisfaction of these equations corresponds to reaching an extremum of $\min_{\varepsilon(t)} J$, and rather general considerations suggest that there will likely be many solutions in typical realistic applications [26]. Each solution would correspond to a particular laser field $\varepsilon(t)$, producing a locally

optimal control outcome. In practice, finding a local optimum is quite satisfactory, provided that $\langle\psi(T)|O|\psi(T)\rangle$ is sufficiently close to the target value.

Many field designs have been performed with optimal control theory for broad classes of applications [22] including rotational [27], vibrational [4, 28], electronic [5], and reactive processes [29]. Notwithstanding the success at achieving designs, the significant computational tasks posed by Eq. (4) have confined the performance of such designs to relatively simple models of molecules or systems with a modest number of quantum states. Even more fundamentally, quantitative knowledge of H_0 and μ is seriously lacking in the most interesting cases (e.g., often the more complex molecules). These comments translate into serious numerical and theoretical challenges, especially considering that successful control in the laboratory will likely draw on high-finesse constructive and destructive quantum wave interferences. These latter interference processes may be rather intolerant to control field designs that are contaminated by uncertainty in the system Hamiltonian and significant numerical approximations in solving the design equations. Nevertheless, approximate designs can be of high value to reveal information about the nature of control fields and provide physical insight into the control of quantum phenomena [30]. Furthermore, the design limitations are not fundamental, and the tasks should be amenable to algorithmic advances, albeit perhaps slowly, in the coming years.

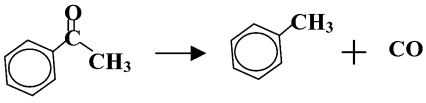
The strongest evidence for the value of performing optimal control designs is provided by the foundation they established for laying out how to perform the analogous optimal control experiments (OCE) directly in the laboratory [18, 19]. Thus, the following section will consider the execution of OCE [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 24].

3 Performance of optimal control experiments

The many successful numerical studies involving the iterative solution of the design equations (Eq. 4) led to the suggestion [18, 19] of leapfrogging the theoretical design process to go directly into the laboratory and perform OCE. Theoretical studies [4, 5, 21, 22] indicated that successful laser field $\varepsilon(t)$ designs would likely have a high degree of temporal or frequency structure, reflecting the fact that the optimal fields for best meeting the objective need to "take over" the dynamics and manipulate all relevant modes or degrees of freedom. This statement is also consistent with quantum systems typically having broadband multifrequency dynamics. Coincident with these observations was the development of ultrafast laser sources, and especially the ability to shape the laser fields in keeping with the requirements of theoretical control designs [31]. All of these points drawn together led to the very simple laboratory architecture [18, 19] for performing OCE shown in Fig. 1.

Figure 1 indicates that a theoretical design could be used as an input trial field, but virtually all of the current experiments were performed with just random input fields [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 20]. Such seemingly poor quality input trials can be rapidly improved upon by taking advantage of the high throughput nature of the experiments, whereby millions may be performed on the order of minutes, and the learning algorithms in the loop often can be rather rudimentary and still recognize the essential features in a rapidly evolving sequence of experiments. The OCE process circumvents the theoretical and computation difficulties stated in Sect. 2 by drawing on the fact that a system subjected to control "knows" its full Hamiltonian with no uncertainties and "solves" its own Schrödinger equation in real time (i.e., as fast as it possibly can be done!) with full precision [18, 19].

Many successful OCE have been carried out in systems ranging from atoms up to complex molecules and solid-state materials [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16]. An illustration of one application is shown in Fig. 2 for the dissociative rearrangement of acetophenone to form toluene [6, 7].



(5)

The OCE apparatus deduced the appropriate laser field to achieve this task in a matter of minutes in the laboratory without any prior design estimate. The field intensity of the original unshaped laser pulse was quite high (around 5×10^{13} W/cm^2), suggesting an overall mechanism whereby the laser field introduced many photons into the molecule to excite a broad set of rovibronic states in just the appropriate fashion to yield

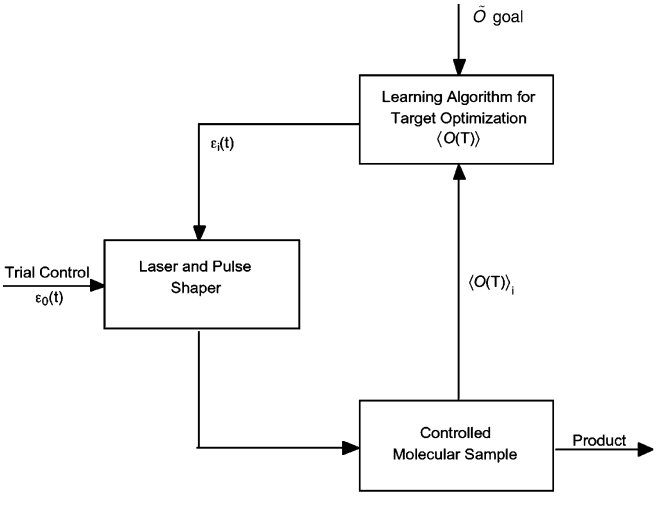


Fig. 1. Closed-loop learning process for performing optimal control experiments (*OCE*). The process is initiated by specification of a product goal $\tilde{O}$ for the expectation value $\langle O(T)\rangle$ of the target operator O at the final time T. A trial field $\varepsilon_0(t)$ from optimal control theory or another estimation procedure may be fed to the laser pulse shaper for the first control experiment. In a sequence of excursions $i = 1,2,...$ around the loop, the learning algorithm guides the shaped laser pulse to steer $\langle O(T)\rangle$ toward the product goal $\tilde{O}$ by observing the patterns of behavior evident in the laser control settings for $\varepsilon_i(t)$ and their molecular actions $\langle O(T)\rangle_i$

the desired outcome. Furthermore, at these field intensities, significant dynamic power-broadening occurs, smearing out the detailed structure in the field-free Hamiltonian H_0. At first sight, it might appear that the latter high-field smearing effects would be counterproductive for delicate control applications; however, operating in this regime can be very attractive, especially for overcoming the limitation of requiring a precise match between the molecular energy level spacings and the laser carrier frequency (recall the original thinking that the precise monochromatic nature of lasers would be a desirable feature for achieving molecular manipulations). The advantages of operating in the strong-field regime for molecular control [6, 7] are indicated in Fig. 3. In this regime, the simple Hamiltonian structure in Eqs. (1) and (4) may need modification, as severe electronic polarization may occur during the control process.

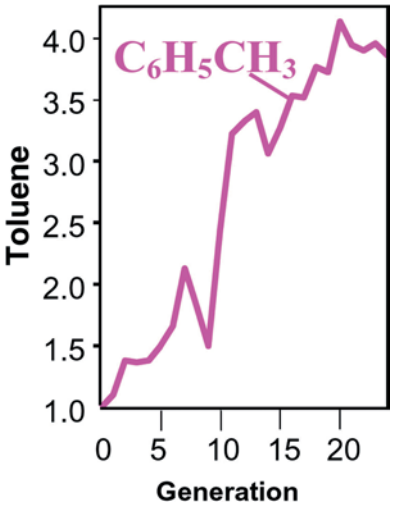


Fig. 2. The average signal for toluene as a function of generation when maximization of the signal for this reaction product was specified for optimization [6, 7]. The parent molecule is acetophenone, where the OCE process in Fig. 1 learns how to tailor a laser pulse to form the toluene product as indicated in Eq. (5)

The operation of OCE often starts with random controls, producing initial data on the scope of the accessible product states, followed by specification of one state for closed-loop optimization. The learning algorithm guides the subsequent sequence of experiments, dictating a continual reshaping of the laser pulse until a maximum yield is attained in the desired state. The present studies [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 20, 24] indicate that OCE are capable of achieving dynamical manipulation of broad classes of systems, including those possibly of high complexity [15].

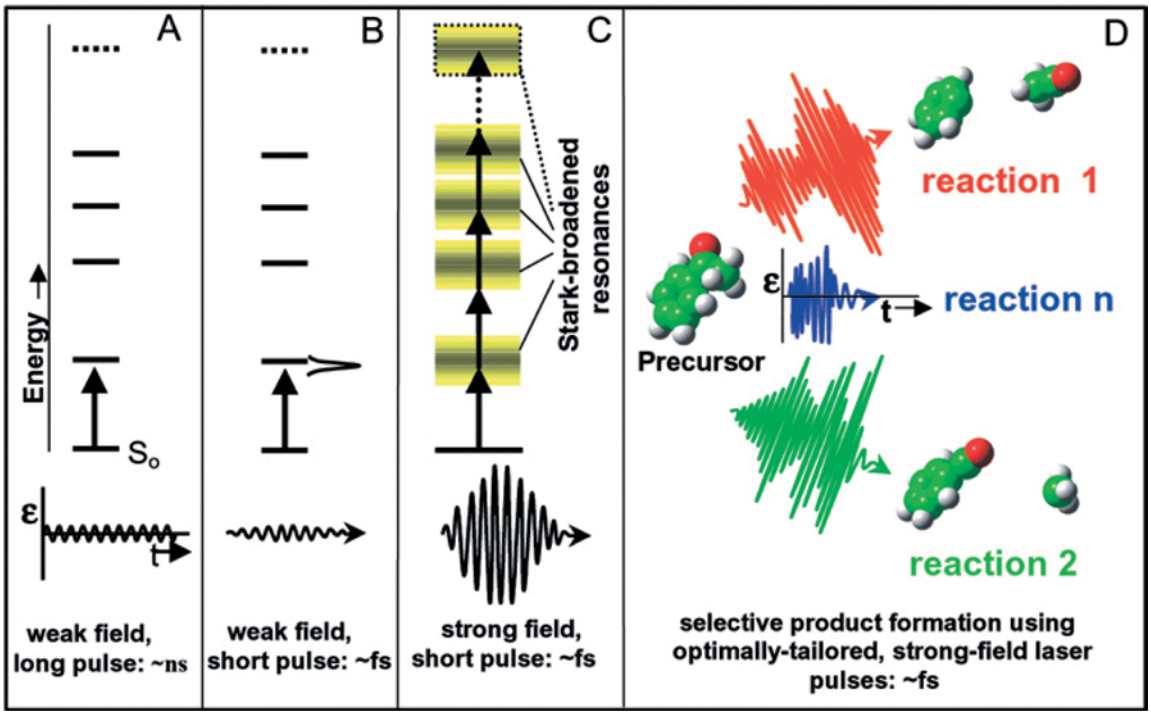


Fig. 3. Laser pulse schemes for creating coherent molecular motion with sources of ever-increasing bandwidth and selectivity capabilities [6, 7]. In each case, the precursor molecule begins in the ground vibronic state, S_0, and interacts with the electric field $\varepsilon(t)$. *Panel a*: In the weak-field case, the molecule can only respond to the laser if the radiation is in resonance with the relevant molecular levels. *Panel b*: In this case, the shorter duration of the pulse helps to overcome resonance constraints. *Panel c*: The high-intensity, short duration of the pulse creates sufficient dynamic power broadening of the molecular vibronic levels (denoted by *yellow bands*) and multiphoton excitation to overcome any restrictions on resonant excitation. *Panel d*: This case indicates how optimal tailoring of the pulse in *panel c* can be used to steer the molecular dynamics out one reaction channel versus another for high product selectivity. Controlled dynamics results from a combination of time-dependent ultrafast multiphoton excitation and Stark broadening of the molecular eigenstates. Selectivity is achieved by a particular intense laser pulse shape stimulating the necessary molecular dynamics to favor a desired product channel

An important feature of performing OCE is the high duty cycle of traversing the loop in Fig. 1. Presently, this duty cycle is limited by the time it takes the laser pulse shapers to switch from one independent shape to another [31]. Many experiments use liquid-crystal and acousto-optic pulse shapers. Besides the switching rates, other operating features of pulse shapers can dictate which type is most appropriate for a particular application. Signal averaging must also be performed on each cycle around the loop. Nevertheless, in a short period of perhaps minutes, it is feasible to perform around 10^6 independent experiments with shaped laser pulses. This number could rise even further with suitable engineering permitting the lasers to operate for extended periods in a stable fashion. Although the full capabilities of OCE for controlling quantum phenomena have not been explored at this time, it is now possible to perform what may be called laser-driven combinatorial laser chemistry and physics [32]. This attractive capability might seem to obviate the need for further theoretical activities in order to achieve successful control of quantum systems. However, this perspective is shortsighted and does not consider the many other issues and applications that lie ahead [33]. Thus, the next section aims to explain some future roles of theory in this field.

4 Theoretical challenges ahead

Projecting into the future in any area is difficult, and the discussion in this section will only attempt to point out broad areas that may particularly benefit from theoretical contributions and guidance. A more focused set of open questions for theory in the pursuit of quantum control is also available [33]. In considering the future role of theory, a most important factor is the advancing OCE capabilities [18, 19, 20] for performing high duty cycle experiments, as sketched out in Fig. 1. The only other area where an analogous capability exists is in combinatorial chemistry, and the optimal control laser experiments have a far higher duty cycle than in (wet) combinatorial chemistry [34]. Bio-combinatorial chemistry occurring *in vivo* appears to operate at high duty cycles with massive parallelization. There surely will be applications of laser control to simple systems where enough information is available to perform an *a priori* theoretical field design as a forerunner to an experiment; however, such cases are likely to be in the minority, given the desire to control complex materials and polyatomic molecules, including those of biological relevance [15]. Fortunately, theory, in this domain, can serve many purposes beyond the design of control fields. The remainder of this section will present a set of specifically identified topics, with the aim of indicating how theory may play a role in addressing the challenges ahead. In many cases, theoretical studies may be enabling for the sought-after laboratory and conceptual advances. The material here is not exhaustive in this regard, but rather indicative of the very positive contributions that theory can make.

4.1 Design and modeling

The computational execution of control field designs, notwithstanding all of the difficulties involved, led to the algorithm for performing OCE [18, 19]. A central issue in the performance of OCE is the nature of the cost function guiding the learning algorithm [20]. The cost function is the experimental analog of J' in Eq. (2) for theoretical design, but in this case, the laboratory cost function can only contain quantities that may be directly observed. These quantities include the electric field and any accessible observables available by ultrafast optical, or other, detection means. Laboratory cost functions minimally must contain sufficient information to guide the experiments to meet the physically posed goal regarding the target operator O. Other competing processes associated with additional observable operators O' could be included, along with possibly further criteria, to meet ancillary needs. The identification of viable laboratory cost functions could be greatly aided by simulating control experiments on judiciously chosen physical systems that are amenable to reliable OCT modeling. The use of simple systems should often not be a hindrance, as the point of such studies is to establish the feasibility of performing a new class of experiments, rather than predicting their precise outcome. The flexibility in choosing cost function structure should evolve as additional experimental capabilities emerge.

Beyond performing theoretical designs for closed-loop algorithm guidance, there is significant need to generate quantum dynamical models, even of a qualitative nature, to describe complex systems under control. A special circumstance is the treatment of control in the high-field regime where molecular rearrangement and possibly ionization occur [7, 35]. Simple theoretical considerations rooted in the Born–Oppenheimer approximation suggest that the field may first induce electronic excitation that subsequently couples to the nuclear motion for molecular rearrangement of various types. In a broader context, modeling of virtually all aspects of quantum control would be valuable to gain physical insight into the control processes. Once again, in many cases, qualitative or semiquantitative models may be quite adequate. The development of reliable means to even approximately solve the design equations (Eq. 4) for complex systems could also significantly advance the field.

4.2 Control mechanism identification

The modeling activities indicated previously may reveal the general features of controlled quantum dynamics, but they would not likely identify the precise mechanisms operative in any particular system of even modest complexity. The power to carry out high duty cycle closed-loop control experiments [18, 19, 20] also suggests the means to identify mechanisms. Once a successful control field has been obtained, large numbers of variations or sensitivity analyses around that field could be performed, along with their observed system

responses. The goal is to make this process systematic and as quantitative as possible in order to extract mechanism information. Theory could be invaluable in this regard, to indicate how to perform the experiments and extract their encoded information [36]. Little precedent exists for specifying the necessary procedures, but one situation to avoid is the need for high-quality offline simulations to extract the desired mechanism information. The algorithms guiding the experiments to reveal quantum control mechanisms would best operate by exploiting the ability of the chemical/physical system under control to solve its own Schrödinger equation, as already utilized on in the OCE themselves [18].

4.3 Hamiltonian feature identification

Although OCE can successfully operate virtually free of Hamiltonian information, theoretical design and analysis requires knowledge of the system Hamiltonian. Hamiltonian information is also valuable for many applications beyond control itself. The extraction of Hamiltonian details (i.e., potential surfaces and optical coupling coefficients) from laboratory data has been a longstanding activity. The tasks involved produce an inverse problem, but one of a far more demanding nature than typically posed by optimal control alone. In quantum optimal control processes, the effort would be considered successful, if any field is found that meets the objectives to a respectable degree. In contrast, when seeking Hamiltonian information from laboratory data, we desire to find the true Hamiltonian.

The optimal-control-type experiments offer a new capability for obtaining Hamiltonian information through the introduction of tailored control fields [37]. In this case, the fields would not be introduced to reach a particular physical target state, but rather to yield the best data for optimally identifying the Hamiltonian. Unlike OCE, the "target" Hamiltonian would not be known beforehand. A set of closed-loop experiments may be considered where the goal is to achieve the most well-defined (i.e., narrowest) distribution of Hamiltonians consistent with the data [37]. Many theoretical and experimental issues need to be considered in the development of viable closed-loop procedures for this purpose. The raw capability of applying many laser fields followed by observations will likely serve little purpose without the appropriate algorithmic guidance on which experiments to perform and how to extract their underlying information content. Simulations of this overall process, even on relatively simple systems, could be valuable for establishing the algorithms for the subsequent execution of the experiments and the extraction of their Hamiltonian information.

5 Conclusion

This paper aimed to present various theoretical considerations in the rapidly developing field of control over quantum phenomena. From one perspective, the exploration of quantum system control may be viewed as old, with nearly a 40-year history going back to the earliest days of laser development. But, the real excitement in the field lies in research initiated in recent years. The successful experiments [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 24] and the capabilities of the laboratory tools [18, 19, 20] suggest that many interesting advances lie ahead in controlling quantum phenomena. The subject deserves exploration for the fundamental insights it may give into quantum dynamics, and it is anticipated that practical applications may also arise from these efforts. Theory and experiment need to be close partners in order to see these developments through to fruition. The best utilization of theory will likely occur in those areas that take direct advantage of the unique ability to perform massive numbers of automated control experiments. The challenges ahead are significant, but they are amply matched by the benefits that may accrue from bringing quantum phenomena under control.

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