EDITORIAL

Focus on quantum control

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Abstract. Control of quantum phenomena has grown from a dream to a burgeoning field encompassing wide-ranging experimental and theoretical activities. Theoretical research in this area primarily concerns identification of the principles for controlling quantum phenomena, the exploration of new experimental applications and the development of associated operational algorithms to guide such experiments. Recent experiments with adaptive feedback control span many applications including selective excitation, wave packet engineering and control in the presence of complex environments. Practical procedures are also being developed to execute real-time feedback control considering the resultant back action on the quantum system. This focus issue includes papers covering many of the latest advances in the field.

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1. Quantum control through Hamiltonian engineering

Much of what we know about science at the atomic and molecular scales comes from various spectroscopies, whose initial studies date back more than a hundred years. In spectroscopy, the general paradigm is to apply weak radiation in order to produce a pristine fingerprint of the undistorted system. In contrast, the field of quantum control seeks to actively manipulate dynamical processes at the atomic scale with systems in the gaseous or condensed phases. The primary means for executing such control is through the application of tailored strongly interacting electromagnetic fields. In recent years, the technology in this area has focused on employing suitably shaped laser pulses, which can orchestrate the quantum dynamics of the electrons and atoms in a sample to attain states or products that would not ordinarily be accessible. Thus, the general paradigm for quantum control rests on engineering the system’s Hamiltonian in order to guide the ensuing quantum dynamics phenomena. The power of at will manipulation of atomic scale Hamiltonians opens up almost limitless fundamental investigations and potential practical applications. The history of this subject can be traced back to at least the 1960s at the time of the invention of the laser. The modern era of the field over the last two decades has witnessed dramatic changes through the introduction of appropriate theoretical and experimental tools leading to the establishment of the key principles of quantum control and a rapidly growing number of demonstrations of the concepts. This focus issue presents a cross section of many recent advances. These latter developments are separated into theoretical and experimental components, but in some cases these designated labels blur as theoretical analyses are often performed to guide laboratory studies while some experiments are done to provide theoretical insights. This synergism is the hallmark of the quantum control field.

2. Recent developments: theoretical

The theoretical study of quantum control encompasses many subjects, including mathematical and computational analyses with an emphasis on the optimal design of laser pulses to manipulate model systems. Much physical insight has been gained from these studies, and the recent efforts are proving to be most valuable. A particular topic of interest is the impact upon control behavior of field constraints, as inevitably arise in the laboratory. Optimized laser fields may still be effective for control even with reasonable constraints [1]. An additional issue in the laboratory is operation with significant uncertainties (e.g. lack of full knowledge about the system Hamiltonian, inhomogeneities in the sample, and statistical variations in the applied controls). Robust controls may mitigate against these factors, and in some cases it appears possible to obtain high quality control along with robustness [2, 3]. The modeling of quantum control often considers Hamiltonians that are linear in the applied field, which couples into the system through the electric dipole operator. However, at high field intensities nonlinear effects can directly enter starting with a quadratic field dependence through the electric polarizability. Control field design in this domain involves additional complexities and trajectory-tracking techniques can be effective [4]. Commonly accessible ultrafast laser techniques can produce pulse trains including those consisting of half-cycle pulses. Controls of this form repeatedly ‘hammer’ on a quantum system in a prescribed fashion, and a number of applications are amenable to control through this process such as tailored excitation and dissociation [5] as well as various nonlinear spectroscopies [6]. Although the control of quantum systems isolated from
their surroundings provides an ideal physical picture, in all realistic experiments the quantum system interacts with an environment of some form. Typical cases involve the quantum system residing in a solvent or host lattice with the system–environment coupling possibly playing a significant role in the ensuing dynamics. The modeling of controlled quantum dynamics in an environment poses challenging demands, and relatively few studies of this sort have been carried out. The formulation of such controlled dynamics requires the development of viable approximate models [7], and time dependent Hartree–Fock techniques are suitable for treating an environment of many identical particles [8]. The control of molecular rotation, alignment and orientation attracted early attention in the quantum control field, and recent years have seen a resurgence of activity in this area with experimental successes. Much still remains to be explored including the prospect of rotating molecules clockwise or counterclockwise by suitably tailored ultrashort laser pulses with spatial polarization [9]. It is also possible to combine the capabilities of ultrafast optimally shaped laser pulses along with an applied dc field to create high degrees of orientation [10]. Additionally, special controls for producing orientation may also be utilized to induce the molecules to become a source of pulsed THz radiation for use in other applications [11]. A promising direction is the application of laser pulses with polarization shaping to atoms and molecules bound to nanostructures containing metallic components. In this case, the laser field’s spatiotemporal shape can depend on the local environment of the nanostructure, and proper optimization of the applied field as well as the nanostructure can create unique opportunities for control [12]. Most control experiments involve an adaptive feedback process, whereby a new sample of quantum systems is introduced on each cycle of the feedback loop with the goal of homing in on the proper optimal control field in an iterative fashion. In contrast, real-time feedback control aims to follow a single quantum system through a sequence of control experiments in a feedback loop. In this case, the feedback process may involve either active measurements with real-time computation to design an updated control [13] or continuous correction of the system’s dynamics by linking it to an auxiliary system serving as a real-time controller. Experiments of this type are a significant challenge, and theoretical insights can be valuable for providing guidance [14]. Although traditional bench top lasers are capable of creating field strengths directly competitive with those that bind electrons to atoms, there is a continuing effort to develop lasers with increasing intensity. The resultant high fields could open up possibilities for producing extreme nonlinear optical effects and even directly manipulating nuclear reactions. An understanding of the basic physics of these controlled processes is at an early stage of research in keeping with the evolving laboratory capabilities [15].

3. Recent developments: experimental

Intense laser pulses were available in the nascent period of laser development and the femtosecond regime was also broached many years ago. With these resources, the application of a simple unshaped intense laser pulse to any sample of matter can readily produce dramatic and permanent changes including high degrees of excitation, ionization, bond breaking, etc. Notwithstanding the transformations produced through application of such brute force, experiments of this type are not normally categorized as examples of achieving control. Under these conditions, there are few variables to serve as active controls to manipulate the induced dynamical processes. The advent of ultrafast laser pulse shaping changed this situation with routine applications now involving upwards of nearly a thousand pulse shape control variables.
for optimizing the resultant quantum dynamical outcomes. In this regard, one goal is selective vibrational excitation on the ground electronic state of molecules with lasers operating in the mid-IR. Suitable shaping of these pulses can lead to controlled vibrational dynamics for a wide variety of applications [16]. In some circumstances, selective vibrational excitation may be used to discriminate for the presence of one species over that of a background of similar ones [17]. These latter studies generate the required complex laser pulse waveforms through adaptive feedback control guided by evolutionary algorithms. Other techniques for achieving control are also available, including the manipulation of quantum mechanical interferences along two pathways leading to photoionization and dissociation. In this case, the spatial phase of the laser can play a special role as demonstrated in experiments on acetone and dimethyl sulfide [18]. There is much interest in the detailed nature of the quantum coherences generated by ultrafast pulses interacting with a sample. In this regard control-probe experiments can often exhibit oscillatory signals, which may reflect either true quantum mechanical interferences or possibly intra-pulse interferences masking the quantum effects [19]. The sculpting of atomic wave packets provides a clear example of manipulating quantum dynamics phenomena, and operation with exited Rydberg states can lead to a detailed understanding of the controlled dynamics. Such a demonstration has been provided upon creating excited Ba$^+$ ng states [20]. Photoionization is often either a direct goal or one of secondary accompaniment resulting from the application of strong laser pulses to atoms and molecules. It is now common to observe the resultant positive ions, and further insight can be obtained from the simultaneous observation of the ejected electrons. Chirped femtosecond laser pulses have been applied to sodium atoms in various excited states for this purpose along with a thorough theoretical analysis of the experiments [21]. The algorithms guiding adaptive feedback control experiments function by observing the patterns of the evolving phase and amplitude variables of a pulse shaper. In many cases, the desired product can be optimized to high yield without specific knowledge of the actual control field. There is much interest in accelerating the process of learning the optimal control field as well as fundamentally understanding the dynamical mechanisms induced by the control, and both goals may be aided by suitable representations of the fields. Control fields can be expressed in the time or frequency domains, and the richest flexibility arises in dual time–frequency representations of fields [22]. Two photon processes have proved to be a basic training ground for quantum control experiments, including the physical insights gained due to their ease of modeling. Much remains to be learned from these simple nonlinear control problems, and new field representations are proving to be helpful [23].

4. Future developments

Research attempting to control quantum phenomena has been underway for some 50 years, but in many respects the field could be viewed as a few years young with the basic physical principles only now beginning to reveal themselves. An important challenge is to explain why optimally controlling quantum phenomena is relatively easy to attain in the laboratory. The understanding of this behavior appears to lie in the topology of the control landscape defined by the observable as a function of the control variables [24]. The control landscape topology has a remarkably simple generic form, and the full implications of this finding remains to be explored. A prime focus of current laboratory studies is on control at femtosecond time and Angstrom length scales. This regime fits many molecular and condensed phase applications, which are readily accessible with the Ti:sapphire laser. New laser resources operating in the attosecond
domain are also becoming available. Dynamics at this scale directly addresses electron motion, and control in this domain will surely be of increasing attention in the future. There are serious challenges in this regard, including for the generation of suitable pulse shapes. In parallel to operating at ever shorter timescales is the desire for achieving control at ultrashort length scales, ultimately in the atomic nucleus. In this case, advances towards increasing laser intensity will be important. Thus, one can envision the full scope of quantum control as eventually ranging over many orders of magnitude of length and timescales. Despite the evident limitations of currently available laser resources, they have already enabled the successful control of many types of quantum phenomena. Each advance in control resources is expected to open up significant new realms for quantum control and enhance the quality of the achieved results. The recent advances in the quantum control field emerged through joint efforts from the theoretical and experimental communities, and this cooperation is expected to be crucial to the future development of the field.

5. Conclusion

The wide diversity of research represented in this focus issue clearly shows the scope of the quantum control field along with a glimpse of what can be imagined in the future.

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