REVIEW: CHEMISTRY

Whither the Future of Controlling Quantum Phenomena?

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This review puts into perspective the present state and prospects for controlling quantum phenomena in atoms and molecules. The topics considered include the nature of physical and chemical control objectives, the development of possible quantum control rules of thumb, the theoretical design of controls and their laboratory realization, quantum learning and feedback control in the laboratory, bulk media influences, and the ability to utilize coherent quantum manipulation as a means for extracting microscopic information. The preview of the field presented here suggests that important advances in the control of molecules and the capability of learning about molecular interactions may be reached through the application of emerging theoretical concepts and laboratory technologies.

Recent years have witnessed rapid acceleration of research activity in the general area of controlling quantum phenomena (*1*, *2*). Most applications have considered laser control over electronic, atomic, and molecular motion, and the roots of the topic may be traced back to the earliest days of laser development in the 1960s. From its inception to the present time, the field of coherent control has been stimulated by the objective of selectively breaking and making chemical bonds in polyatomic molecules. Ultrafast radiative excitation of chemical bonds is often a spatially limited, local process on a molecular scale, and desirable reactive processes may occur before the spoiling redistribution of the excitation into other molecular modes takes place. Under special circumstances, local excitation may be achieved by simply exploiting favorable kinematic atomic mass differences, but this is not a broadly applicable approach. The promise of coherent control, and especially its optimal formulation, is to create just the right quantum interferences to guide the molecule to the desired product.

After many years of frustrating theoretical and laboratory efforts, a recent burst of activity promises advances in controlling quantum phenomena. The present rejuvenation of the field is due to a confluence of factors. These include the establishment of a firm conceptual foundation for the field, the introduction of rigorous control theory tools, the availability of femtosecond laser pulse–shaping capabilities, and the application of algorithms for closed-loop learning control directly in the laboratory. The serendipitous marriage of concepts and techniques stimulated this article, which takes a distinct perspective as a preview, rather than a review, of the field of the coherent control of quantum phenomena.

The resurgence of activity aimed at controlling quantum systems is motivated by the unusual products or phenomena that may become accessible. The long-standing goal of creating novel stable or metastable molecules still is an important objective. For example, ozone has been predicted to exist in a ring configuration of high energy content (*3*), but conventional photochemistry preparation techniques have failed to create this molecule in the gas phase. Another example is the isomerization of acetylene to form vinylidene, a reagent of considerable chemical interest (*4*). The hope in such cases is that the manipulation of molecular motion–induced quantum interferences will open up products or molecular states that are not easily attainable by conventional chemical or photochemical means. Laser-driven coherently controlled molecular dynamics is envisioned to have special capabilities for coaxing the atoms into forming the desired products. Although successful demonstrations of coherent laser control have at long last been performed, the challenge ahead is to fully demonstrate the superior capabilities of coherent molecular manipulation.

One significant recent change in the field is the rapid growth in the type of applications of controlled quantum phenomena being considered, including their use for control of electron transport in semiconductors and the creation of quantum computers (*5*). Although these directions are distinct from the original goal of controlling chemical reactions (*1*), a most important point is that the operating principle for quantum control of any type is the manipulation of constructive and destructive quantum mechanical interferences. Application of this principle can be viewed as a microworld extension of the traditional double-slit wave interference experiment, and the concept has been demonstrated many times in recent years, with an illustration of two-photon absorption in Na shown in Fig. 1, A and B. In this case, the analog of interference produced by the slits is now created by interfering the different pathways $(\omega_1 + \omega_2)$ and $(\omega_1' + \omega_2')$ between the initial (3s) and final (5s) atomic states. The paths may combine either constructively to maximize the absorption signal (Fig. 1B, left) or destructively to minimize the signal (Fig. 1B, right). In more general applications in molecules, achieving high-quality control that cleanly discriminates among the accessible final product states naturally demands playing on multiple molecular "slits" (that is, many interfering pathways) by introducing flexible laser controls that can fully cooperate with the dynamic capabilities of the molecule. This calls for a short laser pulse where the phase and amplitude of the frequency components are controllable. As the principles and techniques are often quite similar in the different applications of controlled quantum phenomena, this preview focuses on molecular applications.

Researchers are also beginning to look beyond the goals of manipulating microworld events as the prime objective. Many of the fundamental contributions of the subject may arise from the finessed quantum manipulation of molecules producing a unique source of data to learn more reliably about the most intimate atomic-scale interactions. Coherent molecular motion depends sensitively on intramolecular forces, and this sensitivity is reflected in the detailed structure of the experimental data. Observations of such dynamics should be a rich source of data for inversion to learn about interatomic forces.

This review explores issues relevant to molecular control through an expression of the current state of the field, an enumeration of desired physical objectives, and an assessment of the techniques necessary to meet the objectives. As a preview of the expansion of the coherent control field through the use of ultrafast laser pulses, the main focus is on the issues and techniques that need attention for the creative evolution of the subject.

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Physical-Chemical Quantum Control Objectives

There are two general reasons for considering coherent control of quantum phenomena: first, to create a particular product that is unattainable by conventional chemical or photochemical means and, second, to achieve a better fundamental understanding of atoms and molecules and their interactions. With these goals in mind, many two-pathway experiments (*6*, *7*) and some pulse control studies (*8*–*11*) have been successfully applied in a variety of quantum systems. Two-pathway experiments have established the validity of the underlying principles (*6*, *7*) and are capable of achieving effective control in appropriate circumstances. However, the number of possible control objectives and systems open to two-pathway control appears limited, compared with what may be attained through the coherent manipulation of quantum dynamics by means of ultrashort pulses.

Current experimental and theoretical studies suggest that large classes of molecules may be brought under coherent photodissociative control, but the possibility of chemical production by this means may remain elusive for a variety of reasons, including merely the cost of the photons per mole involved. Many other molecular applications are also worthy of pursuit, including the cooling of molecules to very low temperatures with tailored laser fields (*12*). Besides the objectives of creating unusual products and molecular states, the interrogation of the ensuing coherent dynamics may provide a new class of data with special capabilities for learning about atomic and molecular interactions (see below). Many laboratory techniques and theoretical concepts will need to be interactively incorporated for coherent control to be broadly successful.

Theoretical Design of Quantum Controls

Theory is playing a major role in conceptualizing the principles of coherent control (*1*, *2*, *13*–*16*), as well as providing designs for the control fields to manipulate molecules in the laboratory (*15*–*17*). Except for the simplest applications, intuition alone as a means of coherent control "design" generally will fail because the manipulation of constructive and destructive quantum wave interferences can be a subtle process. At the other extreme is the application of rigorous optimal control theory design techniques that can find the best way to achieve the molecular objective by manipulating interferences. The procedure is to iteratively solve the Schrödinger equation describing the molecular motion and thereby to optimize the form of the laser field so as to best meet the molecular manipulation objective.

Until now, control field design has largely focused on numerical explorations of possible control fields and associated algorithms to accelerate the design process. Special challenges include finding designs in the intense field regime and for cases in which incoherence effects occur because of molecular collisions or spontaneous radiative emission. The goal of optimal control is to provide laser designs for transfer into the laboratory to implement with the molecular sample. The ability to obtain high-quality laser control designs is hampered by the lack of complete information about the interatomic forces and the computational challenges of solving the nonlinear design equations. Therefore, an increased emphasis is being placed on providing control field estimates whose application in the laboratory would at least give a minimal signal in the target state for refinement of the closed loop, as described in the next section.

Quantum Learning and Feedback Control

A significant advance toward achieving control over quantum phenomena was the suggestion to introduce closed-loop techniques in the laboratory (*18*). The need for such procedures stems from the inability to determine reliable laser control designs, especially for polyatomic molecules. There are two general forms of laboratory closed-loop controls: learning control (*18*) and feedback control (*19*). Learning control involves a closed-loop operation where each cycle of the loop is executed with a new molecular sample; feedback control follows the same sample from initiation of control to the final target objective. These two incarnations have distinct characteristics.

The overall components of closed-loop learning control in quantum systems are depicted in Fig. 2. The procedure generally involves three elements: (i) an input trial control laser design, (ii) the laboratory generation of the control that is applied to the sample and subsequently observed for its impact, and (iii) a learning algorithm that considers the prior experiments and suggests the form of the next control for an excursion around the loop once again, and so on. If the molecular control objective is well defined, the control laser of appropriate capability, and the learning algorithm sufficiently intelligent, then this cyclic process will converge on the objective. In some cases, a quasi-random field might be used (that is, the investigator goes in blind) to first detect an initial weak signal from the product molecule for further amplification. In especially complex cases, it may be necessary not only to detect the product but also to observe critical intermediate molecular states in order to efficiently guide the closed-loop process toward convergence. It should be noted that the laser field need not be measured in this learning process, as any systematic characterization of the control "knobs" will suffice. This procedure naturally incorporates any laboratory constraints on the controls, and only those pathways to products that are sufficiently robust to random disturbances will be identified. Full computer control of the overall process is essential, as the loop may be traversed thousands of times or more in order to converge on the molecular objective. A variety of learning algorithms may be utilized for this purpose; presently, genetic-type algorithms are being employed, although others have been considered in simulations. More attention needs to be given to the nature of these algorithms and, especially, their stability in the presence of lab-

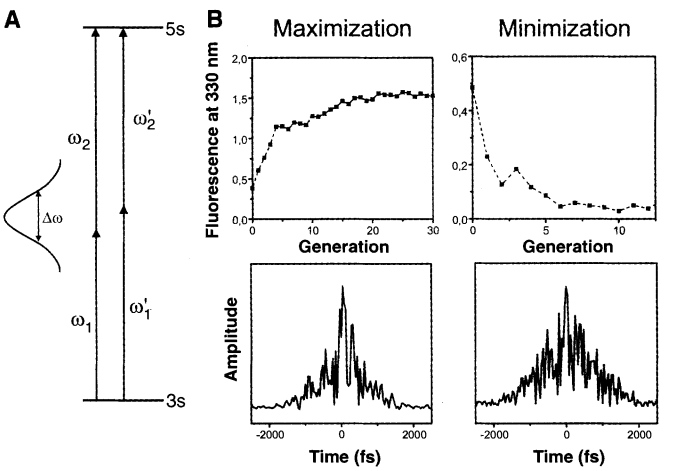

Fig. 1. (**A**) The fundamental principle of coherent control, realized with the two-photon transition 3s → 5s in sodium (*8*). The upper level is excited by pairs of photons when the sum of their energies (for example, $\omega_1 + \omega_2$ and $\omega_1' + \omega_2'$) equals the two-photon resonance. Because a femtosecond pulse of width $\Delta\omega$ is spectrally broad, many distinct pairs exist, representing different paths. If the phase between the different paths is appropriately chosen, then constructive or destructive interference occurs leading to maximization or minimization of the two-photon transition probability. The femtosecond laser can be tailored to modulate the phases using a pulse shaper controlled by a learning algorithm, as shown in Fig. 2. The 330-nm fluorescence of Na(4p) → Na(3s) is used as the feedback signal to the algorithm. (**B**) The data for fluorescence maximization on the left and minimization on the right. The upper panels show typical learning control convergence curves for the two different cases when evolutionary algorithms are applied. The lower panels show the envelopes of the electric fields of the two optimized phase-shaped pulses.

oratory disturbances. Typical quantum control objectives are expected to have multiple successful laser control solutions, and it is to be hoped that at least one may be found in the learning process.

Toward Combinatorial Laser Chemistry

Recent experiments with closed-loop learning control have been successful going in blind for the manipulation of fluorescence signals, photodissociation products, and other applications (*8*–*11*). An illustration of photodissociation control is shown in Fig. 3 (*10*). In this case, the objective was to break selected bonds of the $CpFe(CO)_2Cl$ molecule (here, Cp denotes the cyclopentadienyl ion) by either maximizing the product $CpFeCOCl^+/FeCl^+$ branching ratio (Fig. 3, black bars) or alternatively minimizing the same ratio (Fig. 3, white bars). A distinct optimal laser field was found for each of the two processes with laser phase modulation techniques, guided by a genetic learning algorithm. Experiments such as these could be referred to as combinatorial laser chemistry, by analogy with similar quasi-random chemical synthesis techniques. It is expected that the successful control of virtually all quantum dynamics phenomena, especially of a complex nature, will require the use of closed-loop learning techniques.

By repeatedly starting with a new molecular sample, learning control sidesteps the issue of whether the observation process may disturb the subsequent evolution of the quantum system. In contrast, feedback control would work with the same quantum system from time of control initiation through evolution to the final state. In some cases, feedback might be introduced for reasons of stabilization around a nominal operating condition, as perhaps, in a quantum computer. Limitations on the speed of electronics and the manipulation of general electro-optical elements suggest that quantum feedback control experiments will likely apply to systems having long natural time scales. Overall, a diagram similar to that in Fig. 2 is also operative for quantum feedback control; however, some distinct differences exist. First, the feedback algorithm must operate with sufficiently reliable knowledge of the system Hamiltonian to make good judgments on the subsequent controls while the quantum system is still continuing to evolve. Second, a clear competition exists between the desire to use the control algorithm to get as much information as possible about the evolving state of the quantum system, and the fact that gathering ever more precise information will increasingly disturb the system and lead to a larger degree of uncertainty about its control. Although some mathematical analysis of feedback control has been considered (*19*), the practical feasibility of quantum feedback control remains an open question, especially for cases involving many cycles around the control loop. As a first step toward feedback experiments, the continuous observation of an evolving quantum state has been made (*20*). For control objectives that are not overly demanding, it is possible to consider the performance of observations that minimally disturb the system. An example might be the control of a Bose-Einstein condensate cloud of atoms where weak observations of only a scattered few atoms should be sufficient to characterize the control of the overall spatial features of the cloud. Molecular motion is also often semiclassical, suggesting the existence of a broad class of feedback control behavior lying somewhere between the strict limitations of quantum mechanics and the less demanding situation found in classical engineering applications.

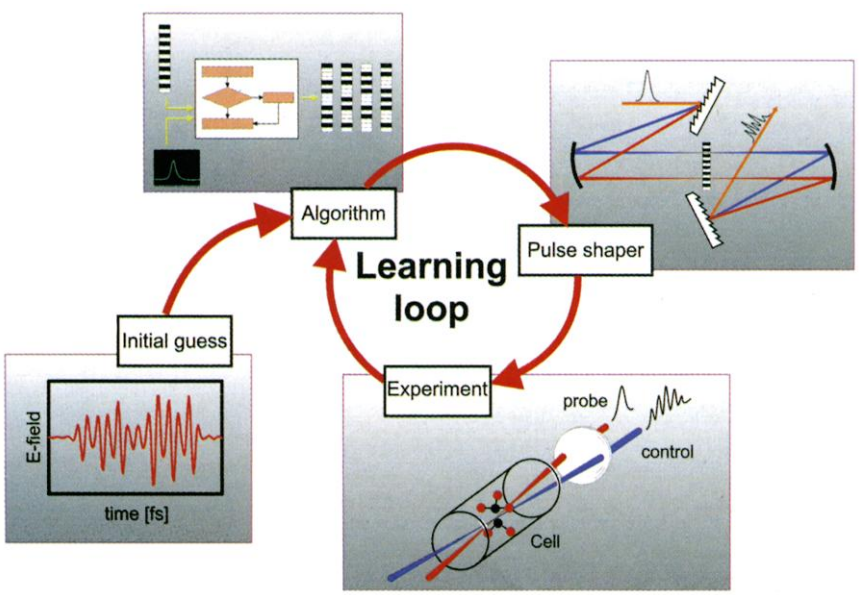

Fig. 2. A closed-loop process for teaching a laser to control quantum systems. The loop is entered with either an initial design estimate or even a random field in some cases. A current laser control field design is created with a pulse shaper and then applied to the sample. The action of the control is assessed, and the results are fed to a learning algorithm to suggest an improved field design for repeated excursions around the loop until the objective is satisfactorily achieved.

Laboratory Realization of Quantum Controls

The laboratory implementation of molecular control has been demonstrated with continuous wave lasers (*6*, *7*) and, in the time domain, with ultrashort laser pulses (*9*, *10*, *21*). Although the use of continuous wave lasers is effective in some cases, in general, a few frequencies will not be sufficient to create the necessary interferences in complex problems where a number of quantum states have to be simultaneously brought into constructive superposition. Pulses lasting less than 20 fs correspond to a bandwidth of about 1000 cm^{-1} and, therefore, provide many independent laser frequency subcomponents that can be tuned simultaneously with a pulse shaper (*22*). Because no electronic device is fast enough to work on a femtosecond time scale, only frequency domain laser pulse–shaping is feasible. In the near-infrared (IR) spectral range, this technique is well developed for laser phase and amplitude modulation. Acousto-optical modulators (AOM) (*23*), as well as liquid crystal modulators (LCM) (*24*), have been used for modulation, and LCMs are commercially available for the spectral range from 430 nm to 1.6 μm.

The AOM combines the advantages of high spectral resolution and fast response times, but it also suffers from low transmission of the laser light (typically, 5%) and more complex implementation. The LCM exhibits high transmission (~80%) and easy implementation, but it has low spectral resolution (typically, 128 discrete pixels) and slow response times of at least a millisecond (*23*, *24*). Fast response times are important for closed-loop laboratory optimization algorithms, as many thousands of distinct shapes need to be produced, along with repeated experiments for signal averaging.

Control of molecules in their ground electronic state is attractive for many applications, and for this purpose, it is necessary to develop pulse shapers that work in the mid-IR and far-IR spectral ranges (*25*). Breaking this problem down into parts, possibly around 3 μm, 5 μm, or 10 μm, would allow for the manipulation of fundamental motions in many compounds (for example, the C–H stretch, and torsional and skeletal motions of

organic molecules). Mid-IR femtosecond pulse energies of ~10 μJ with temporal pulse lengths of ~200 fs are already available, and they should be sufficient for pulse shaping and manipulation of highly excited molecular vibrations. The extension to longer wavelengths, improvement of tunability, and increased intensities, as well as the generation of very short femtosecond pulses in these spectral ranges, are all important for practical applications. The development of new types of nonlinear crystals already shows promising results in those directions. An open question concerns the ability to lock the phases of two laser pulses with different frequencies in order to coherently manipulate different vibrations in the molecule (*26*). The phase-locking would be inherent if it becomes possible to create ultrashort pulses in the IR.

The quality of current laser pulse-to-pulse stability appears to be adequate for many applications. The presence of modest noise can even help the search for a better solution in a complex multidimensional control field search with closed-loop laboratory learning algorithms (*27*). Certain applications (for example, quantum computation or those working in the high-intensity regime) may be more sensitive to laser fluctuations, and these cases need further evaluation.

Bulk Media Influences

The current molecular closed-loop control experiments (*10*) have focused on the idealized situation of isolated molecules in a low-density gas or molecular beam. However, many important chemical, physical, and certainly biological processes take place in the condensed phase. Operating in this regime (*9*) raises questions about the effects of the surrounding media on the coherent processes. These issues break into two categories of media effects on the molecules under control and on the propagating control field. All media effects may not be bad, as a molecule under control could benefit from the restraint of the atomic motion by the immediate surrounding medium, thereby providing partial dynamic guidance toward the target. A naturally occurring example of this phenomena exists in the photoisomerization of rhodopsin as a part of the visual process. In this case, the protein matrix in which the rhodopsin is embedded reduces the number of possible degrees of freedom for the outcome of the photoisomerization step to produce an efficient and selective visual excitation signal. On the other hand, the same media interactions may lead to the transfer of the controlled excitations of the target molecules to the media and a consequential loss of control. At present, little is known about these processes in the coherent control regime. Furthermore, the medium can influence the propagating laser pulse even in the gas phase, if the cell length is sufficiently long. A passing laser pulse will polarize the medium, which, in turn, can provide feedback and reshape the pulse even if there is no absorption. This effect can be serious for control applications, as the delicate phase structure of the pulse is very important for quantum manipulation. However, theoretical studies have indicated the possibility of using laboratory learning control techniques to identify the laser pulses for transformation of the entire bulk medium as best as possible.

Quantum Control Rules of Thumb

With successful coherent control experiments now emerging in the gas and condensed phases, it is time to consider whether some consistent rules for achieving coherent control can be found. Chemistry operates with a relatively small number of rules, supported on a rational physical basis, prescribing the properties of a vast ever-growing number of molecules and materials. These rules serve to guide chemists in endeavors ranging from ab initio electronic structure studies to the synthesis of complex molecules. An open question is whether the domain of coherent control may similarly be reduced to a set of logical rules to guide successful manipulation of quantum phenomena. A comparison with traditional chemistry operating in the incoherent regime suggests that the new rules might be easily revealed owing to the more ordered conditions in coherent control. However, no such coherent control rules have emerged, except for the simplest notions of working with molecular absorption selection rules and spectral resonances in the weak field regime. Coherent control rules could be envisioned to range from the identification of common mechanistic pathways to characteristic control features associated with distinct classes of molecular objectives. Insight into this matter may be revealed by examination of the structure of the control fields, as well as consideration of the overall family of successful control solutions observed in the laboratory. Operating in the intense field regime opens up the prospect of making up the rules as desired, because the molecule and its electronic orbitals could be severely distorted at will. Seeking control rules of thumb also raises questions about how to categorize dynamic similarities and differences between molecules, as well as for control fields. Although an eventual lack of coherent control rules of thumb would not diminish the significance of the subject, establishing such rules would make the control of new systems easier to achieve. The best evidence for rules is likely to emerge only from the observed trends in the control of large numbers of chemical and physical processes.

Extracting Microscopic Information from Coherent Dynamics

In going beyond the use of molecular excitation for control purposes, the observation of controlled dynamics is also rich in information on the underlying interatomic forces. Learning about interatomic forces has been a long-standing objective in the chemical sciences, and extracting that information from observed coherent dynamics entails finding the appropriate data inversion algorithms for this purpose.

Various forms of continuous wave spectroscopy have been the traditional source of data for attempts at extracting intramolecular potential information. Although such spectra are relatively easy to obtain, serious algorithmic problems have limited their inversion to primarily diatomic molecules or certain special circumstances with polyatomics. This traditional approach has many difficulties, including the need to assign the spectral lines and to deal effectively with the inversion instabilities. As an alternative, coherent control techniques may be used to launch a molecular excitation to scout out portions of the molecule and to report the information back by probing with ultrafast lasers.

The second approach lends itself to arguments that experiments in the time domain may provide the proper data to stabilize the inversion process. These arguments generally reduce to the use of localized wave packets as a means of achieving stabilization. Although any single experimental observation would not yield global-scale molecular information, it may unambiguously identify a local region. It appears that the ultimate source of data with this goal in mind are "movies" of atomic motion obtained from ultrafast imaging techniques. A theoretical analysis (*28*) indicates that such data would be ideal, as it admits an inversion algorithm with unusually attractive characteristics, including (i) no need to solve

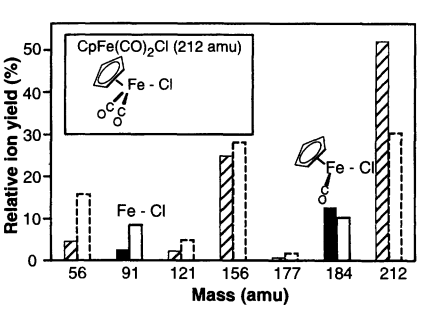

Fig. 3. The controlled fragmentation of $CpFe(CO)_2Cl$ achieved with a femtosecond laser pulse shaped in a phase modulator (*10*). The ratio of $FeCl^+$ (mass 91) to $CpFeCOCl^+$ (mass 184) production is either maximized (black bars) or minimized (white bars) by the use of the closed-loop learning process depicted in Fig. 2. The figure also shows additional uncontrolled fragments from the molecule as dashed and cross-hatched bars.

Schrödinger's equation, (ii) a rigorously linear mathematical formulation calling for no iteration for data inversion, (iii) inherent stability arising from the imaging data explicitly defining the region of the potential that may be reliably identified, and (iv) no need to know the control pulse initiating the dynamics. Although it is early to speculate when such molecular movies (*29*) of high resolution and quantitative accuracy will be available, these algorithmic considerations provide strong motivation for pushing in this direction. In addition, the utility of other types of coherent temporal data for inversion needs consideration.

Conclusion

Significant advances have been made in recent years toward establishing the broad foundations and laboratory implementation of control over quantum phenomena. Since its inception, this field has anticipated the promise of success just around the corner, but now, there are preliminary experimental successes. Another important new feature is the increasing breadth of controlled quantum phenomena being considered, and success in one area will continue to foster developments in others. A basic question is whether molecular control executed in the coherent regime offers any special advantages (such as, new products or better performance) over working in the fully incoherent kinetic regime. Answering this question should be a major goal for the field. However, even in cases where incoherent conditions are best, the same systematic control logic may produce better solutions than those attainable by intuition alone. Although creating unusual molecular states, or even functional quantum machines (for example, quantum computers), could have great impact, the ultimate implications for controlling quantum processes may reside in the fundamental information extracted from the observations about the interactions of atoms. The synergism of femtosecond laser pulse-shaping capabilities, laboratory closed-loop learning algorithms, and control theory concepts now provides the basis to fulfill the promise of coherent control.

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