Quantum Physics Under Control

Thanks to the increasing ability to coherently control quantum systems, designer Hamiltonians can be created to explore new physics and to yield a better understanding of complex phenomena.

Ian Walmsley and Herschel Rabitz

The preparation, controlled evolution, and measurement of specific quantum states are fundamental activities in physics. The study of new states of matter and the new perspectives on quantum physics that are provided by processing the information are but two of the important reasons for pursuing such research. The numerous potential applications range from performing precision measurements to manipulating molecular nanoscale devices. Optimally designing the control and measurement strategies is important for extracting the most information about the state of the system from a given set of measurements. Tools based on control theory have been developed for such purposes for systems that obey the laws of classical physics. But for quantum systems, redirecting those tools—and possibly introducing new ones as needed—is a challenge.

That control concepts may be useful for robustly creating particular quantum states has long been recognized. The development of complex pulse sequences in nuclear magnetic resonance is a widely exploited example. Since the invention of lasers some 40 years ago, a goal has been to achieve laser-selective molecular transformations based on the controlled deposition of energy within molecules (see PHYSICS TODAY's special issue on laser chemistry, November 1980). These applications and others rely on the coherent evolution of quantum systems to achieve the desired manipulations.

Being able to use tailored external fields to freely manipulate quantum systems has significant implications for physics, and concepts of control can lead to practical realizations of that goal in the laboratory. The number of successful quantum control experiments is rising rapidly, although the research area is still in its infancy. Many issues remain, including the degree to which quantum systems may be controlled, the identification of the best tools for their manipulation, and the nature of new physics that may be discovered from applying quantum control concepts. Such ideas have had a long gestation period in the physics community, and this article summarizes the current status of the field.

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Controlling quantum phenomena

Quantum control refers to active intervention in a system's dynamics to maximize the probability that the system evolves toward a desired target state (see figure 1). For example, quantum control might be used to redirect a chemical reaction along a specific pathway, or to precisely operate a quantum logic gate in the presence of environmental noise. In addition to producing the desired evolution, control of quantum systems promises to provide a refined means for learning about the behavior of the systems themselves.

Quantum systems subjected to control range from a single atom or molecule to the collective degrees of freedom, such as excitons and phonons, in solids. A common tool for exercising control is the optical field from a laser. Shaped ultrashort laser pulses are proving to be very versatile, because the time-dependent amplitudes and phases of the pulsed fields can be tuned to match the multiple frequencies of the electronic and vibrational degrees of freedom of atoms, molecules, and excitations in solids.[1]

One can control a quantum system's degrees of freedom either directly or indirectly. When a laser pulse is used to control a molecule, for example, the electromagnetic field may drive electronic excitations through a dipole interaction; depending on the couplings of the electrons and nuclei, those excitations may be transferred to the molecule's vibrational motion or to the spin degrees of freedom of nuclei and other electrons. Lasers can also directly control other degrees of freedom, such as the vibrational motion of molecules and crystals, by appropriate choice of the optical wavelength (typically in the infrared).

Most quantum systems have a rich spectral structure, and the target state typically is a superposition of many eigenstates (as would be the case for spatial or momentum localization of the excitation, for example). Optical fields that are spectrally broad are thus often the most useful. The different spectral components must be coherent, however, because the coherence of the light is mapped onto the quantum coherence of the system under control. Pulses from lasers satisfy these requirements; moreover, they can be shaped with great precision (see the box on page 44). Dramatic progress in laser technology over the past decade has made possible almost arbitrary control over pulse shapes at the femtosecond level, with wide wavelength flexibility.

The general protocol for achieving quantum control is,

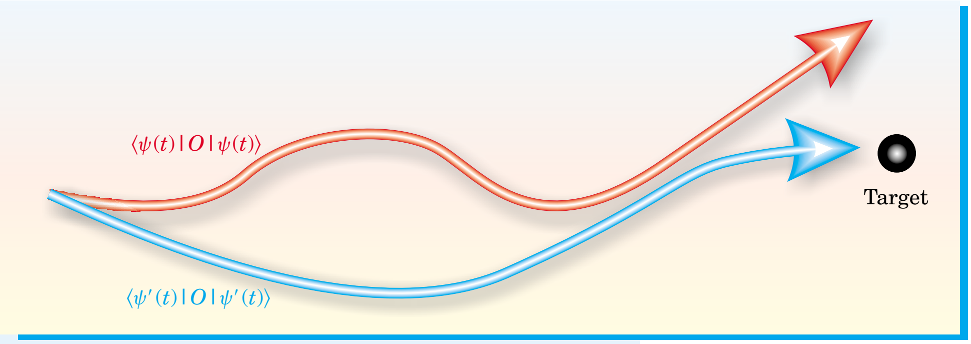


Figure 1. The influence of control on quantum dynamical evolution. Without control, a system's wavefunction $\psi(t)$ will evolve under the free Hamiltonian H_0, and the expectation value of the observable operator misses the target. Under controlled dynamics, an additional, externally controlled term in the Hamiltonian alters the evolution of the wavefunction $\psi'(t)$ and directs the expectation value of O to evolve toward the target.

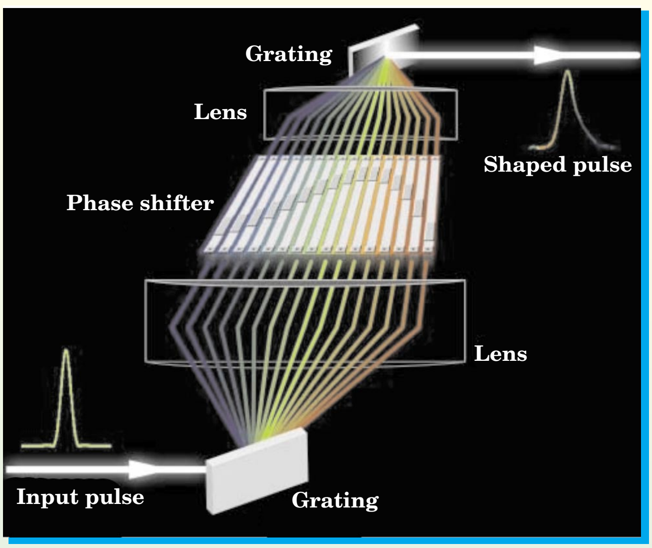
in principle, straightforward: Introduce a trial control and measure the system performance using a sensor; then modify the control to correct for deviations from achieving a designated target. Such a protocol is termed closed-loop control. A closed-loop experiment begins by selecting a trial pulse shape to drive the quantum system. There are typically a large number of input control settings—for example, the amplitudes and phases of each of the spectral components of the optical control field. After applying the control pulse, one observes the quantum system, perhaps by measuring the absorption of an ancillary short pulse or by probing the dynamical nonlinear optical susceptibility of the system. A merit function measures the quality of the achieved control and provides a basis for determining how to modify the shape of the next trial laser pulse. In this way, a closed-loop search for the control settings maximizes the merit function while taking into consideration experimental constraints, such as fixed pulse energy, maximum pulse intensity, or the need to not destroy the quantum system under study!

Taking a control perspective in physics is quite natural. For instance, virtually all experimenters endeavor to optimize a particular signal, and methods of control provide a framework for experimental design and operation to achieve that goal. Control will likely become a necessary part of the increasingly complex experiments undertaken today. For example, stable operation of multivariable systems such as quantum information processors will probably require real-time feedback and a closed loop that "teaches" the control system—a process called learning control. Tolerance to uncertainties such as environmental effects can be part of the experimental design from the outset, provided the Hamiltonian is known to sufficient precision. Construction errors or Hamiltonian uncertainties inevitably arise in manipulating large systems at atomic and larger scales, yet learning control is capable of optimizing the system performance a posteriori, regardless of the errors.

Physics is fundamentally about understanding, and a profound reason for adopting control methods is their ability to provide a better means to understand how quantum systems operate. Achieving control requires relating the target state to the control as it enters the system Hamiltonian; one can thus, in principle, learn about the system by combining an understanding of its controller with observations of the resulting dynamics. Data inversion algorithms, for example, can direct a control experiment toward robustly determining Hamiltonian parameters such

Shaping laser pulses

Ultrashort optical pulses can be shaped by adjusting the phase and amplitude of each spectral component. The most common schemes for this use a Fourier-plane shaper.[17] In that device, the input pulse is incident on a grating that disperses the different colors in different directions, as shown in the figure. The colors are collimated and focused by a lens or mirror. A second similar arrangement in reverse reconstitutes the pulse by redirecting the colors to another grating. At the mutual focal plane of the two lenses, the spectrum of the input pulse is completely resolved so that each spatial location corresponds to a single frequency (or a narrow band). By inserting at this plane a material that causes variations in the phase of each resolved frequency, one can construct a pulse of arbitrary shape, constrained only by the spatial resolution of the arrangement.



The most frequently used devices for adjusting the spectral phases in this manner are a multipixel liquid-crystal array or a broadband acousto-optic modulator, although a deformable mirror may also be used. Using an electrical signal, one can change the effective refractive index of these devices at any given spatial location in the image plane .[18]

An important feature of this technology is the rapidity with which the pulse shape may be changed: Several thousand updates may occur per second. That response rate is well suited for the output of a typical titanium:sapphire laser system, which, with trains of pulses about 30 nm in bandwidth near a mean wavelength of 800 nm, can produce pulses as short as about 30 fs at repetition rates of a few kilohertz. (Figure courtesy of G. Gerber.)

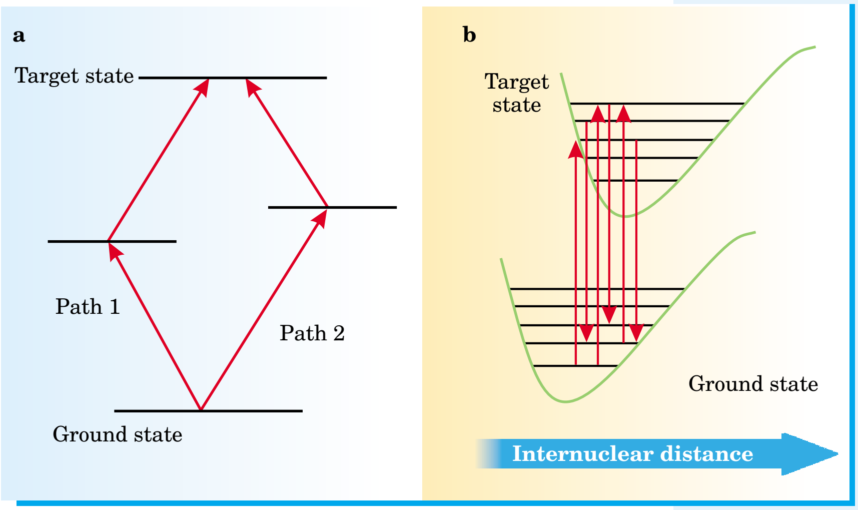


Figure 2. Optical pulses can be used to drive both simple and complex excitations in atoms and molecules. **(a)** Two different quantum pathways link the initial and target states. Optical fields can manipulate the complex amplitudes of the two paths to change the population of the target state. **(b)** A shaped broadband pulse can manipulate many pathways to provide control over more complex dynamics. Shown here are various transitions associated with vibrational motion.

as the electric dipole and electron–vibrational coupling coefficients. It is too early to anticipate all the implications of controlling quantum systems, but the subject should be rich, because controlled manipulations will greatly expand the scope of accessible quantum phenomena.

Achieving quantum control

From a control perspective, two primary features distinguish quantum from classical dynamics: the complexity of solving a system's equations of motion and the nature of the control measurements. In classical physics, the complexity of solving Hamilton's equations of motion scales linearly with the number of particles, whereas for quantum systems, the complexity scales exponentially with the number of particles. That exponential scaling makes designing control fields formidably difficult for anything but the simplest quantum systems. Indeed, a functioning quantum computer would be needed to make the design process truly efficient! In addition, the alteration of a quantum system as a result of a measurement challenges the central tenet of classical control theory—that the system can be observed without disturbing it.

Introducing a control alters the system Hamiltonian, either statically or dynamically. A static control might be a solid-state or molecular structural alteration, whereas dynamical controls, which are more common, involve applying external time-dependent fields such as coherent optical or radio-frequency radiation, or low-frequency electric or magnetic fields. Experiments may simultaneously include both transient and static controls; in some cases, the two classes of controls may be related, because transient fields may induce structural changes.

When a control is applied, the original system Hamiltonian H and its associated quantum dynamics $i\hbar\partial|\psi(t)\rangle/\partial t = H|\psi(t)\rangle$ are altered.[2] The new, full Hamiltonian $H'(t) = H + V(t)$ includes the control interaction $V(t)$—the product of an applied optical field and the electric dipole operators, for example—and redirects the dynamics to the new evolving system state $|\psi'(t)\rangle$. In a control experiment, one typically has a specific target in mind for a specific observable operator O, but the control-free system evolution $\langle\psi(t)|O|\psi(t)\rangle$ does not meet that objective. The goal of control is to direct the new expectation value $\langle\psi'(t)|O|\psi'(t)\rangle$ with the control present toward the desired target at some finite time $t = T$ or asymptotically as $t \to \infty$ (see figure 1).

The conceptually simplest control scheme exploits interference to achieve a specific target. Such a scheme relies on two quantum pathways between the initial and final states and uses the ability to control the phase and amplitude of optical fields, for example, to drive the probability amplitudes of the pathways themselves,[3] as shown schematically in figure 2a. Experiments based on this scheme with one or two control parameters can modulate the target-state yield in quantum systems with few competing dynamical processes. Another approach, the so-called pump–dump, relies on the timing between two laser pulses to manipulate the quantum dynamics such as the fracturing of a specific bond in a triatomic molecule.[4]

Interference, pump–dump, and related techniques typically only have a few control parameters, but they still can be effective in limited circumstances. Interest has rapidly evolved, however, toward the control of highly complex quantum systems, such as polyatomic molecules,[5] biosystems,[6] and quantum information systems. Through the interference of many quantum pathways (figure 2b) in such systems, an external control can access many different outcomes. Thus, a more comprehensive approach to achieving control generally requires a specifically shaped time-dependent control field that strongly drives the quantum system.[1]

In recent years, many control fields have been computed for manipulating various quantum mechanical processes, including rotational, vibrational, and electronic excitation in molecules, chemical reactive processes, solid-state electron dynamics, and molecular laser cooling.[7] Although those designs have yielded much physical insight about controlled quantum dynamics, they are not, for the most part, being implemented in the laboratory. The basic reason is that a successful control generally requires subtle constructive and destructive quantum interferences, which in turn, can depend sensitively on even the smallest details of the system Hamiltonian. But model Hamiltonians typically lack sufficient accuracy, and numerically solving Schrödinger's equation during the design process introduces additional errors. Because of such difficulties, the current quantum control-field designs are, at best, of qualitative reliability, especially for some of the most interesting cases of complex systems and for systems operating in the strong control-field regime. Nevertheless, the quality of control designs is improving, and valuable in-

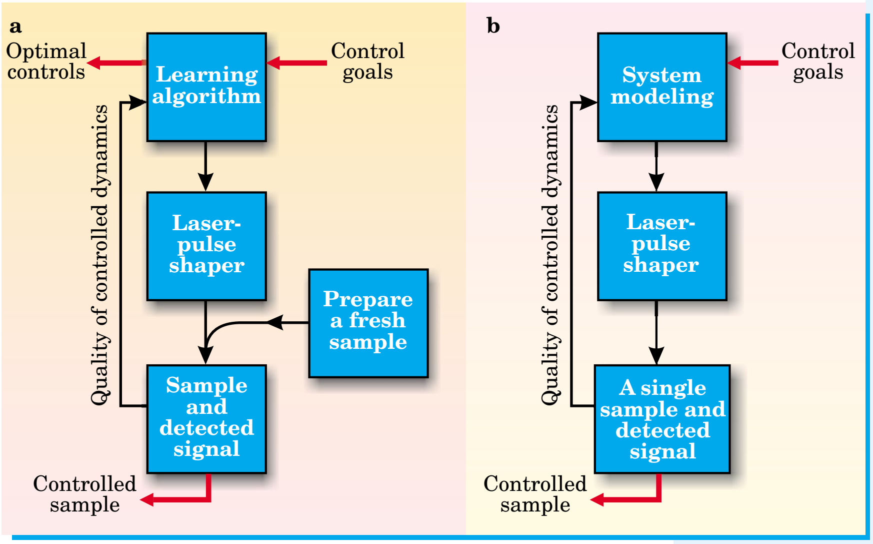


Figure 3. Two schemes for manipulating quantum dynamics phenomena. **(a)** A scheme that uses learned control to optimize tailored laser pulses. The tailored laser pulses induce quantum dynamic excursions in a sample. Under high-duty-cycle closed-loop operation, the process can home in on a particular pulse shape that steers the system as close as possible to the desired target. On each excursion of the loop, a new quantum system is prepared in the same initial state for controlled manipulation. **(b)** A scheme that uses real-time feedback for deducing the evolution of control laser pulses. High-speed modeling or analysis of the quality of the controlled dynamics is used to estimate new controls that minimize dynamical evolution errors, such as those due to environmental fluctuations. Stabilizing quantum system dynamics is a common goal for schemes of this type.

formation will come from further design efforts.

The forward problem of calculating the evolution of a quantum system in state $|\psi'(t)\rangle$ under a prescribed field is linear, because Schrödinger's equation is linear in $|\psi'(t)\rangle$. But the inverse problem of finding the control field that meets a target goal is highly nonlinear, because the target can depend on $V(t)$ in a complicated way. Yet that inverse problem is at the heart of discovering—whether through computational design or directly in the laboratory—effective control fields. In classical engineering control, a premium is placed on modeling the system dynamics because realistic simulations are often feasible and experiments can be very expensive. In the quantum regime, exactly the opposite circumstance is typical: Experiments can be performed much more rapidly, and often more reliably, than simulations.

Carrying out quantum control experiments has become astonishingly easy in cases using shaped pulsed laser fields: Thousands or even millions of independent control experiments can be carried out in minutes. The high duty cycle of laboratory quantum control experiments can be viewed as an automation of a typical experimental protocol: Adjust the apparatus (that is, the controls) to change the experimental conditions in consideration of the system responses. The automation of that process using computer-controlled laser pulse shapers enhances by several orders of magnitude the number of independent quantum control experiments that may be performed; such acceleration is a key feature driving the rapid expansion of the quantum control field and the discovery of new effective controls.

Methods of control

Closed-loop laboratory quantum control is amenable to two classes of experiments: learning control and real-time feedback control. For many quantum systems, observations remove particles or permanently alter their state. In such a situation, learning control takes place on sequential sets of identically prepared systems.[2] In real-time feedback, measurements that only weakly disturb a single evolving quantum system may provide sufficient guidance to reshape the control fields without erasing all information about the state of the system.[8]

In learning-control experiments, the system response typically is recorded after the control field has been turned off, and new systems are prepared in the same initial state for subsequent cycles using different control fields (figure 3a). Repeated iterations lead eventually to an optimized control.[2] Figure 4 shows a typical learning-control experimental setup using a shaped laser pulse for control.

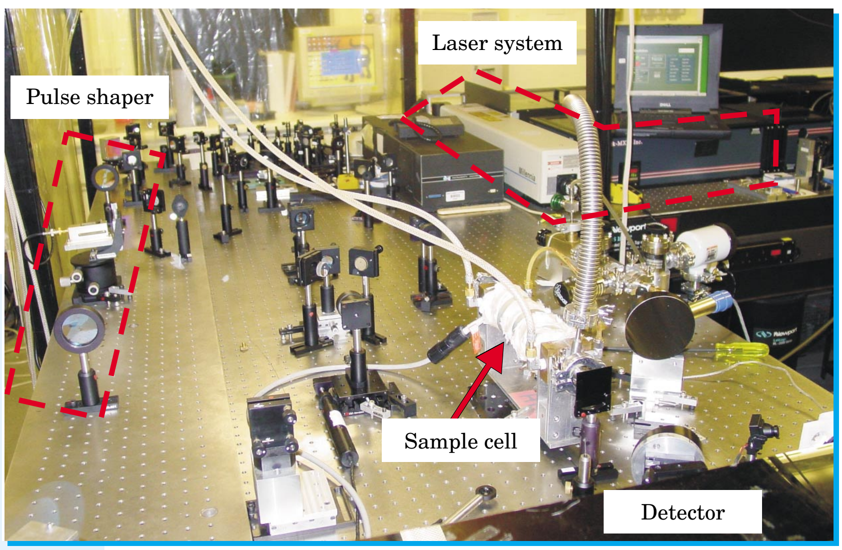


Figure 4. Components of a typical quantum learning-control apparatus include the laser system, pulse shaper, sample cell, and signal detector. Pattern recognition software uses the detected signal to direct the laser-pulse shaper and thereby home in on the desired controlled quantum dynamics of the sample.

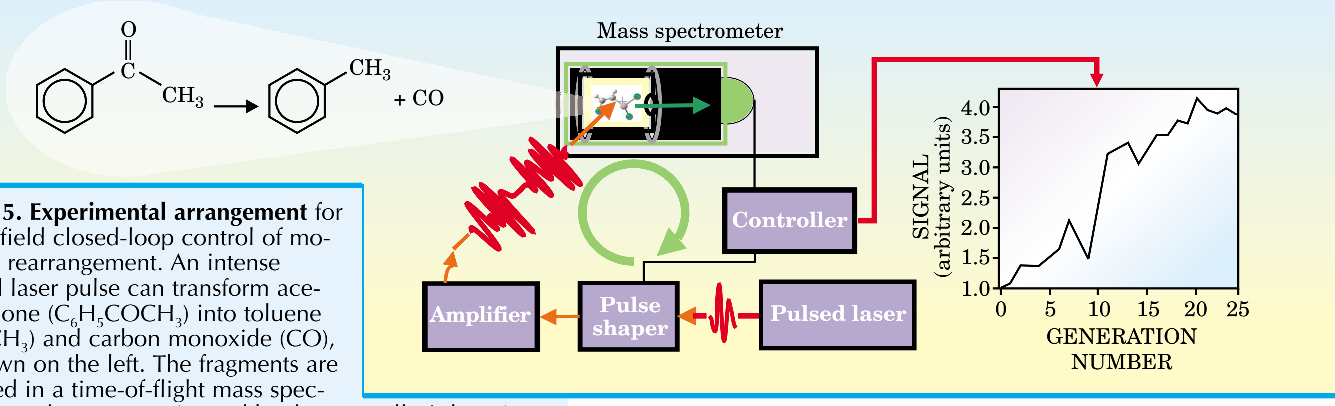


Figure 5. Experimental arrangement for strong-field closed-loop control of molecular rearrangement. An intense shaped laser pulse can transform acetophenone ($C_6H_5COCH_3$) into toluene ($C_6H_5CH_3$) and carbon monoxide (CO), as shown on the left. The fragments are detected in a time-of-flight mass spectrometer, whose output is used by the controller's learning algorithm to optimize the reaction's yield. Over several iterations, the controller directs the generation of a series of shaped pulses until the signal is maximized. The plot shows the average reaction yield as a function of the iteration number for the laser-controlled transformation. The behavior seen in the plot is typical of the learning-control experiments of figure 3a.

Learning control may be viewed as being model-free, because it may be performed with little knowledge of the system Hamiltonian H. The lack of detailed system information is supplanted by performing massive numbers of experiments under the guidance of a pattern-recognition learning algorithm. The repeated experiments systematically explore the relationship between the system response and the settings of the applied control field. A distinct second class of closed-loop experiments uses a sequence of measurements and controls on a single quantum system while it is evolving (figure 3b).[9] That feedback control process typically requires some form of real-time quantum system modeling to redirect the controls as the system evolves. The heavy computational resources needed for modeling may restrict real-time feedback control to simple systems.

The learning-control technique illustrated in figure 3a sidesteps the issue of measurement-induced state changes. Moreover, learning control can operate effectively in the presence of quantum system uncertainties, such as unknown coupling strengths between the control fields and the system states. In addition, averaging the responses of nominally identically controlled systems can incorporate stochastic fluctuations both in the system preparation and in the control field itself; the resulting controlled dynamics can be robust with respect to such noise.

Feedback control also can account for random disturbances during the controlled dynamics and take corrective actions. However, accounting for the influence of the measurement itself on the controlled quantum dynamics is a challenge. Feedback control will always have a degree of latency given by the minimal response time of the apparatus. Such latency is critical when controlling quantum systems, since environmentally induced decoherence of the quantum dynamics may occur on time scales that are comparable with the delays in adjusting the controller. The latency of the feedback loop therefore sets an upper limit on the rate of controlled coherent evolution of a quantum system.

Currently, learning control is used widely in many laboratories,[10,11] whereas feedback control is at an early stage of development. How to combine these two techniques to best control quantum phenomena remains an open question.

In the laboratory

A growing number of successful laboratory experiments use control concepts. Perhaps most significant are the demonstrations of control over strongly driven systems and the experiments—such as the selective dissociation of complex polyatomic molecules—on systems with many coupled degrees of freedom.[12] In certain applications, including modification of chemical reactivity and generation of very high harmonics of a fundamental laser frequency, the information gained from measurements of the control fields and their actions has been used to begin to understand the dynamics of the systems under control.[5,13]

Closing the loop in the learning-control process of figure 3a used to be the job of the researcher (usually a student), who would use the results of one experiment to plan the next on a comfortable human time scale. Now the experiments are so fast that software is needed in addition to the student to recognize patterns and adjust the controls. Currently, so-called genetic algorithms are the popular choice for guiding the sequence of evolving experiments. Such algorithms try a large number of controls and use mutations and combinations of the best settings in the next iteration (termed a generation) to home in on the optimal settings. Figure 5 shows an example of the learning curve for a genetic algorithm.[12] In the first experiment of this kind, Kent Wilson of the University of California, San Diego, and colleagues maximized the fluorescence yield from a laser-excited organic dye molecule.[14]

Among the recent applications of control ideas are the selective dissociation of organometallic molecules targeting a specific ligand for removal[10] and dissociative recombination to selectively break apart a molecule and rearrange some of the fragments (see figure 5).[12] Some experiments are even beginning to reveal how shaped laser pulses can act as a type of fleeting "reagent" to stimulate molecular transformations. Applications involve increasingly complex systems; examples include the manipulation of electronic energy transfer in biological systems,[6] the synthesis of high-harmonic radiation from laser-driven atoms,[13] the creation of an ultrafast solid-state optical switch, and the controlled manipulation of molecular vibrations. These and other experiments are providing tantalizing glimpses of the possibilities for using control tools to alter the outcome of quantum dynamics phenomena.

Beyond the control itself, an important physical goal is to determine the mechanisms that guide the controlled dynamics. The outcome of a nominal experiment contains the inputs needed for that determination: a quantitative measure of how well the desired dynamical objective was met and a record of the control field involved. In a few recent experiments, the control mechanisms have been re-

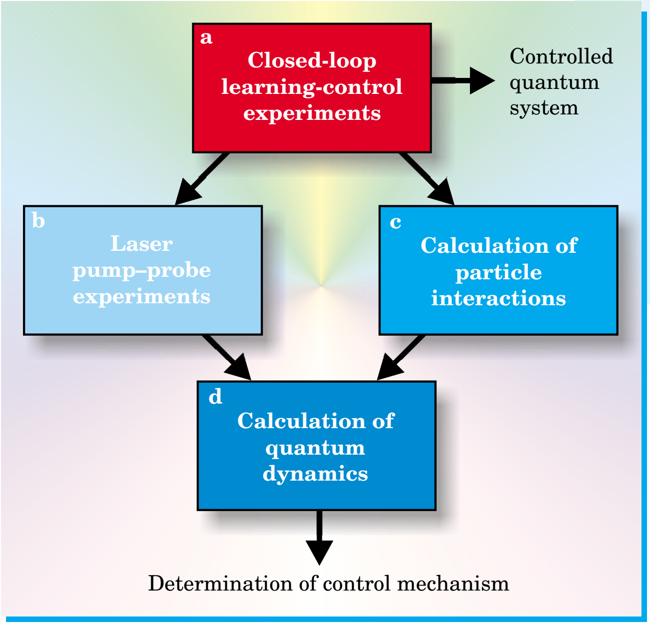


Figure 6. Determining the control mechanism is a second goal of control experiments. As this schematic shows, the closed-loop learning control techniques **(a)** of figure 3a can be combined with additional steps **(b, c, d)** to deduce the control mechanism. Based on information about the optimized control field, extra experiments that use laser pulses to excite and probe the system, along with quantum simulations of the system evolution, can be used to gain insight into the control mechanism.

vealed through dynamic modeling.

One success in identifying the control mechanism occurred in the generation of extreme ultraviolet radiation from scattering intense light pulses off atoms.[13] High-intensity radiation incident on an atom or molecule may kick out an electron, which then will oscillate in the optical field. On each oscillation cycle, the electron might collide and reunite with the parent ion. During the collision, the ion–electron system acts as a nonlinearly oscillating antenna that radiates energy at many—possibly hundreds of—harmonics of the original pulse frequency. The efficiency of that highly nonlinear process is very small, but by using learning control, Margaret Murnane, Henry Kapteyn, and colleagues at the University of Colorado achieved their goal of enhancing a particular harmonic in the extreme ultraviolet (XUV) region. They measured the pulse shape that met that objective and used it in a numerical model of the process. The model revealed that the optimally shaped pulse improved the efficiency of the XUV generation by matching the temporal evolution of the pulse field's phase to that of the ion–electron dipole, whose phase in turn depends on the temporal structure of the pulse. A full numerical simulation of the learning-control experiment produced a control field design and a high harmonic signature similar to those found in the laboratory. The combination of experiments and modeling led to a clear physical picture of the mechanism that produced the selective harmonic generation.

Modeling of the fluorescing dye experiment of Wilson and his colleagues[14] also led to successful identification of the control mechanism. The closed-loop learning-control experiment resulted in a pulse with a large time-dependent frequency shift (a so-called chirped pulse); the direction of the chirp determined the efficiency of the population inversion that is needed for the fluorescence. Jianshu Cao (now at MIT) and his collaborators developed a physical model that explained this pulse shape in terms of vibrational motion of the molecular wavepacket. Their analysis produced a molecular-wavepacket control model that combined the efficiency of electronic excitation with the fidelity of coherent vibrational dynamics.

A recent study by Ludger Wöste and his colleagues at the Free University of Berlin revealed the mechanism through which a control pulse optimized the ionization of an organometallic molecule while reducing competing processes. In that work, the researchers followed a general paradigm, shown in figure 6, that involved additional pump–probe experiments as well as extensive quantum dynamics calculations.[5] Efficiently handling the quantum computations in figure 6 remains a significant hurdle to overcome in attempting to discern the underlying mechanisms of the control experiments.

The degree to which the emerging laboratory control results can be described as either quantum mechanical or coherent is currently unsettled. To the extent that the controls operate on manifestly quantum entities, the results are quantum in character. However, it is not obvious that the results all make full use of quantum interference. In some experiments, for example, the control field can be viewed as manipulating atoms as they move along the so-called vibronic potential energy surfaces (such as those in figure 2b) that correspond to a molecule's vibrational motion;[15] such an interaction may be described classically or semiclassically yet still possibly produce successful levels of control. Because the optical coherence of a control field is initially mapped onto the quantum system, stable broadband radiation sources with well-defined phases between all of the spectral components are critical for creating resultant quantum coherences. Shaping the laser pulses is essential for directing the radiation into the proper states or degrees of freedom in a carefully sequenced or phased manner to manage effectively the ensuing dynamics. And accurate measurement of the pulses is critical for analysis of the dynamics.

Continuing developments in new laser sources should enable even greater degrees of control. It is now possible to set not only the relative phases between different frequency components, but also the absolute frequencies of each component. That capability promises to be an important technology for very broadband control experiments in the attosecond regime, such as the direct control of electron motion in molecules and condensed phases.

What lies ahead

In the quest to transfer control ideas from the familiar realm of classical mechanics to the new opportunities in quantum systems, researchers are already making progress in applying optimized estimation techniques to quantum state tomography—that is, the multidimensional imaging of quantum states. In such applications, ideas from control engineering permit the best estimate of a state or operation to be obtained from a set of measured data and allow the experiment to be designed to ensure that the dataset itself is optimized. In addition, researchers have used optimal filtering for adaptive measurement to detect weak optical fields and to identify Hamiltonians.[8,9] In those applications, the measurement apparatus settings are adjusted as more data is taken, so that the experiment optimally homes in on the value of the desired system parameters.

Quantum control also opens up new possibilities. For example, quantum measurements can project the system being measured into a specific state to reduce the entropy of the system.[16] Eugene Polzik's group at the Niels Bohr Institute in Copenhagen, Denmark, has prepared a highly nonclassical state of many atomic spins by measuring the polarization properties of a probe field passing through the collection of atoms. Mark Raizen, at the University of Texas at Austin, and colleagues have proposed the removal of entropy via measurement as a method of cooling an atomic gas. In their scheme, one would use a measurement made on a small part of the sample to determine a control field to be applied to the entire sample. A sequence of such operations can lead to cooling, but the quantum limits of such control scenarios are unknown.

Science and technology have a symbiotic relationship: Improvements in one often lead to progress in the other. Allying control methods may yield new insights into quantum physics, and new ideas about control may emerge from its conflation with physics. Seth Lloyd of MIT points out that machines imply ideas, and ideas imply machines. The numerous emerging laboratory implementations of closed-loop learning-control methods show that one can successfully tailor exciting radiation to generate selective quantum dynamics. We are just beginning to understand the power of achieving quantum control and the consequences for physics that emerges out of control.

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