# Quantum Control

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**Abstract.** Tunable lasers, sources of intense narrow-band coherent radiation, enabled the parity violation experiments in atoms in the 1970's. Ultrafast lasers are source of *broadband* coherent radiation. This paper reviews recent progress in experiments to control quantum dynamics in condensed phase and gas phase systems, using shaped ultrafast radiation. Many of the same techniques that have led to laser pulses in the 10-100 fsec range can also be applied to the control of quantum systems with similar dynamical time scales

## INTRODUCTION

The following limericks were written during data collection runs on the thallium parity violation experiment in 1977 or 1978:

Commins et al. at UC
Thought they saw a small asymmetry
After sorting out statics
And false systematics
They said it was due to the Z.

The scientists at Novosibirsk
Said "Comrades, we discovered it firsk!
At first we were sad
'Cause our noise was so bad
But compared to yours it look no wirsk!"

Fortson up north in Seattle
Says, "My setup's developed a rattle!
It comes and it goes.
A Z-boson? Who knows?
For weak force, it sure puts up a battle."

And now clear from Oxford we hear
That a change in results may be near.
While the boss was out speaking
The students were tweaking.
Now sometimes strange signals appear.

CP596, *Art and Symmetry in Experimental Physics,* edited by D. Budker et al.

This *festschrift* contribution in honor of the career of Gene Commins is an opportunity to look for connections between those parity violation measurements we all worked on together, and broader and more applied areas of laser science. The 1970s was a decade of great excitement in spectroscopy because of the invention of the tunable laser. As I arrived at Berkeley, Gene's student Steve Chu was designing and building flashlamp pumped dye lasers from parts machined in the shop or scavenged from the Livermore junk pile. The parity violation experiments were an opportunity to apply precision laser spectroscopy to a fundamental problem in physics. Under Gene's mentorship, we learned the joy of this challenge.

The term "quantum control" does not describe a single field of physics, any more than do the terms "laser spectroscopy" or "precision measurements." Rather, quantum control is a method of using laser fields to study different kinds of physical problems. Part of the fun is finding new frontiers where these techniques can make an impact. Examples of these frontier fields are quantum information science and coherent chemistry.

Quantum control is based on the notion that a well-controlled time-varying intense optical field can transform any pure quantum state of a system into a different state with different properties. In some problems, the target state, initial state, and system Hamiltonian are all known, so that the challenge is to get from the start to the finish efficiently. Methods such as Optimal Control Theory have been developed for these situations. Other times, the target state and system Hamiltonian are not known, but some system properties are known. An example is breaking a specific bond in a complex molecule. In this case we cannot prescribe the optimal control field in advance, but the solution can be found through a systematic search. One of our goals has been the construction of a "learning machine," that is, an automated experimental apparatus that can use feedback signals from a quantum system to help optimize control strategies for a particular application. The learning machine consists of a search algorithm, together with programmable experimental inputs and readouts.

## FUNDAMENTALS OF CONTROL

The ideas behind quantum control are illustrated by an examination of the Hamiltonian for a system in the presence of external fields:

$$(H_{System} + H_{External} + H_{Control})\Psi = i\hbar\partial\Psi/\partial t \qquad (1)$$

Here $H_{Systerm}$ is the time-independent Hamiltonian of a quantum system, such as an atom or molecule. $H_{External}$ is the interaction of the quantum system with external fields. These could be due to collisions, or couplings to other modes in the case of a complex molecule, and also include coupling to the vacuum modes of the radiation field that cause fluorescent decay. $H_{Control}$ is the coupling that we add in order to control the system, such as the field of a laser. $\Psi$ is the Schrödinger wave function, and quantum control is the process of directing $\Psi$ into a desired target state.

The evolution of the wave function is governed by the combination of internal and external contributions. In the absence of $H_{External}$, the system has stationary states, the energy eigenstates, and any $\Psi$ can be constructed from a suitable linear combination of these. In real systems $H_{External}$ is never totally absent, however; it leads to loss of the quantum coherences ("$T_2$"processes), and ultimately to relaxation of the excited state amplitudes ("$T_1$" processes).

In most situations in laser spectroscopy, the control Hamiltonian is a small perturbation compared to the external Hamiltonian. In that case only a tiny fraction of probability amplitude can be transferred away from the initial state before relaxation sets in. Quantum control is not possible, since the loss of phase information means loss of control. Control, then, requires strong fields. In the notation of equation (1), we require $H_{Control} > H_{External}$.

Another useful way to look at the control problem is the competition of different time scales in the problem. The control Hamiltonian transfers coherent population among states of the system, a process characterized by the Rabi frequency $\Omega$, which is proportional to $H_{Control}$. The amount of probability amplitude transferred to the excited state goes like $\Omega\tau$, where $\tau$ is the laser pulse duration. Significant population transfers should be much more rapid than the decoherence time, i.e. $\Omega > T_2^{-1}$, and this is equivalent to the strong field control condition expressed above.

Some control experiments can relax this requirement by making use of a "launch state." This is an eigenstate of $H_{System}$ prepared at the beginning of the experiment, which which is relatively immune to $H_{External}$, and thus provides the reference phase for further excitation of a wave packet. Pulsed excitation with a weak field (i.e. where $\Omega\tau<1$ ) can still produce controllable wave packets; however, if we have $\tau > T_2$, then all coherence is lost. This then leads to the two conditions for quantum control: strong fields and ultrafast pulses. Many techniques now exist to produce such optical fields and to shape them. Quantum control has grown out of these advances in technology.

## SHAPING ULTRAFAST OPTICAL PULSES

Ultrafast optical pulses are produced by mode-locked broadband lasers. The mode-locking mechanism is usually a nonlinear focusing element such as a Kerr lens in the cavity, which makes the laser optical cavity most stable if the circulating light is a single very short pulse. If the gain medium is very broadband, the pulse may have only a few optical cycles. We use a Kerr-Lens modelocked titanium sapphire oscillator[1]. The output is amplified in a 10 Hz regenerative chirped pulsed amplifier[2]. The output pulse is approximately 100 fsec long, with a central wavelength of about 790 nm.

The light pulse is then shaped by filtering its spectral components in a zero-dispersion pulse stretcher, consisting of a 1:1 telescope between two diffraction gratings.[3] The heart of this pulse shaper is a programmable acousto-optic modulator (AOM) made of Tellurium Dioxide *($TeO_2$)*, which forms a Fourier filter.[4] The acoustic wave creates a transient transmission grating which diffracts the optical wave at the Bragg angle. Since the acoustic wave is essentially frozen as the optical pulse travels through the crystal, the complex amplitude of the acoustic wave traveling

through the crystal in the transverse direction $A(t)\cos\omega_c t = A(y/v_s)\cos\omega_c t$ is mapped onto the optical field $E(\omega)$ as it passes through the AOM. The shaped beam then has the form

$$E_{shaped}(\omega) = E_{input}(\omega) \times a(\omega) \times e^{i\varphi(\omega)t} \tag{2}$$

where $a(\omega)e^{i\phi(\omega)} = A[y(\omega)/v_s]$.

The shaped pulses can be measured using spectral interferometry. In this technique, the pulse to be measured is combined with an unshaped reference pulse, and then analyzed in a spectrometer. The spectrally resolved interference between signal and reference is a direct measure of the spectral phase function.

## SCULPTED RYDBERG WAVE PACKETS

This section describes our development of wave packet "sculpting" and the quantum interference techniques used to view these sculptures.[5] We use Rydberg atoms to explore ideas in strong field quantum control and quantum information problems. Rydberg atoms are a testing ground for quantum state preparation and measurement.[6]

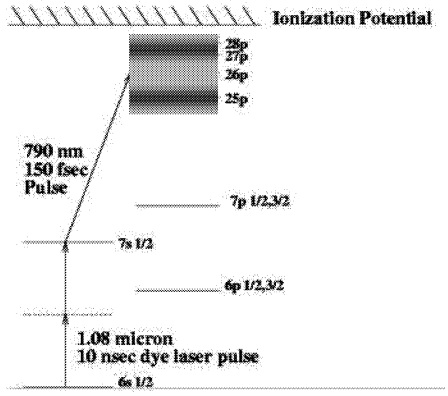


Figure 1. Excitation scheme for producing sculpted Rydberg wave packets in atomic Cs.

Rydberg wave packets are produced in a Cs atomic beam by first exciting ground state atoms to the 7*s* state using a two-photon transition at 1.08 $\mu m$. Sculpted laser pulses then excite the atoms to Rydberg states. This produces coherent superpositions of $p_{1/2}$ and $p_{3/2}$ states with principal quantum numbers between $n$=24 and $n$=35. In most applications we are only interested in tracking the shape over the first few picoseconds, so the spin-orbit interaction (on the order of 100 *ps* for the states considered here) can be neglected. This means that the wave packet can be described without spin, in the *nlm* basis. The value of the magnetic quantum number *m* then

depends on the relative orientation of the quantization axis and the laser polarization that induces transitions from the *7s* state.

Decoherence and dephasing due to Doppler effect and collisions can be reduced by using an atomic beam. Residual dephasing mechanisms are stray electric fields, and some residual first-order Doppler shifts if the beam is not perpendicular to the laser. These contribute to coherent dephasing times on the order of $10^{-7}$s. Decay of the state amplitudes can occur due to excitation by black-body radiation, or fluorescence decay, with associated $T_1$ times on the order of $10^{-5}$s. These times set the limits for producing and measuring wave packets.

The cesium Rydberg wave packet $\Psi(\mathbf{x},t)$ is determined by the amplitude and phase of the constituent eigenstates:

$$\Psi(\mathbf{x},t) = \sum_{np} a_n e^{i\Phi_n} u_{npm}(\mathbf{x}) e^{-i\omega_n t} + a_0 \varphi_s e^{-i\omega_s t}, \tag{3}$$

where $\phi_s$ is the launch state. For weak excitation, we can use Fermi's Golden rule to calculate the coefficients:

$$a_n e^{i\Phi_n} \propto E(\omega_{np} - \omega_{7s}) \langle npm | z | 7s \rangle \propto E(\omega_{np} - \omega_{7s}) / n^3 \tag{4}$$

This expression relates the quantum state amplitude and phase to the amplitude and phase of the sculpted optical field.

## Amplitude Measurement

Quantum amplitudes of Rydberg wave packets are measured using two techniques. The simplest method for atomic beams is the technique of state-selective field ionization[7]. If a uniform electric field is applied to the Rydberg wave packets, field ionization becomes possible. However, the field ionization probability is not a monotonically increasing function of field, because the energy structure of the wave packet in the field is discretized into constituent Stark eigenstates. Each energy eigenstate with energy $E$ has its own characteristic critical ionization field $F$. For alkali atoms, this is roughly given by $|F| = (1/4)E^{-2}$ in atomic units.

When a wave packet experiences the critical field for one of its constituent states, the ionization occurs with probability proportional to the square of the amplitude for that state. Thus, the atomic beam ionization distribution maps the squared state amplitudes.

In some situations, state selective field ionization is inconvenient or impossible. For example, wave packets in a dense gas or a liquid may relax so rapidly that the ramped field technique takes too long, or the ionized electrons cannot be extracted. In that case, it is often possible to use quantum wave packet interferometry to analyze the wave packet amplitudes. Quantum interferometry uses a Michelson interferometer to split the optical pulse that excites the wave packet into two parts with a variable time delay $\tau$. Each pulse in the pair excites an identical wave packet in the atom, and the two wave packets coherently interfere, depending on the time delay:

$$\Psi(\mathbf{x},t,\tau) = e^{i\omega_{gs}\tau} \sum_n a_n u_n(\mathbf{x}) e^{-i\omega_n t} (1 + e^{-i\omega_n \tau}) \tag{5}$$

The total wave function depends only on the squared amplitudes and the time delay:

$$\langle \Psi | \Psi \rangle \propto \sum_n |a_n|^2 \cos \omega_n \tau. \tag{6}$$

A Fourier analysis of the ionization current $|\langle\psi|\psi\rangle|^2$ will yield the amplitudes.

## Phase Measurement

The phase of the state coefficients $a_i$ can be determined using a holographic technique similar to spectral interferometry. In this measurement, the atoms are excited to a second, *reference* wave packet superposition with a time delay $\tau$ with respect to the sculpted packet. The reference excitation has real amplitudes, produced by a transform limited optical field.

$$\Psi_{ref}(\mathbf{x}, t, \tau) = e^{i\omega_{gs}\tau} \sum_n b_n u_n(\mathbf{x}) e^{-i\omega_n t} \tag{7}$$

The probability for state $i$ depends on the relative phase $\phi_t$ between signal and reference

$$P_i = |a_i|^2 + |b_i|^2 + 2|a_i||b_i| \cos[(\omega_i - \omega_{gs})\tau - \phi_i]. \tag{8}$$

Since this delay time $\tau$ is not stable with respect to the optical frequency $\omega_i$-$\omega_{gs}$, the phases are extracted by averaging several laser shots and constructing the correlation function

$$r_{ij} = \frac{\langle P_i P_j \rangle - \langle P_i \rangle \langle P_j \rangle}{(\Delta P_i)(\Delta P_j)} = \cos[(\omega_i - \omega_j) - (\phi_i - \phi_j)]. \tag{9}$$

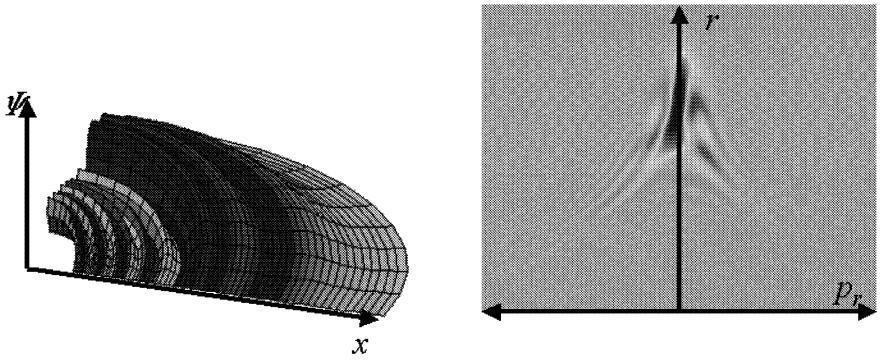


Figure 2. Two views of a sculpted wave packet. Left: Schrödinger wave function. The amplitude is shown as a function of x and z for a *p*-state wave packet oriented along x. phase is represented by the shading of the wave packet. Right: Wigner representation of the same wave packet in the $(r,p_r)$ phase space plane.

# QUANTUM CONTROL APPLICATIONS

## Data Storage and Retrieval

Quantum control techniques can be used to to load, hide, retrieve, and manipulate information in wave packets. Consider, for example, the storage of a simple binary number, such as 000001000. A wave packet could encode this number in various ways. For example, one could load an $n$-state quantum wave packet with an $n$-bit number according to the prescription that at a specified time $t$, the phase is real and positive for binary 0, but real and negative for binary 1. Since every state in the wave packet has equal amplitude, ordinary spectroscopic techniques such as state-selective field ionization cannot reveal which state stores the binary 1. This bit is hidden from view. If the same data were stored in a classical binary register with $n$ locations, one would have to search each location to find the marked bit. The search would take, on average, $n/2$ steps; however, the rules of quantum state manipulation provide some simple methods for revealing the marked bit.

Quantum holographic techniques provide a simple means to find the marked bit. To find the hidden information, the atom is excited by a search pulse which produces a second wave packet with all quantum phases relatively real. This, in fact, is the very same "reference" pulse that was used to measure the phases in the sculpted wave in the previous section. The superposition leads to destructive interference of any states where a binary 0 was stored, but constructive addition to any states with a binary 1. The combined wave packet then has the same information as before, but now it is encoded into quantum amplitudes rather than phases. These can then be read out using state-selective field ionization.[8]

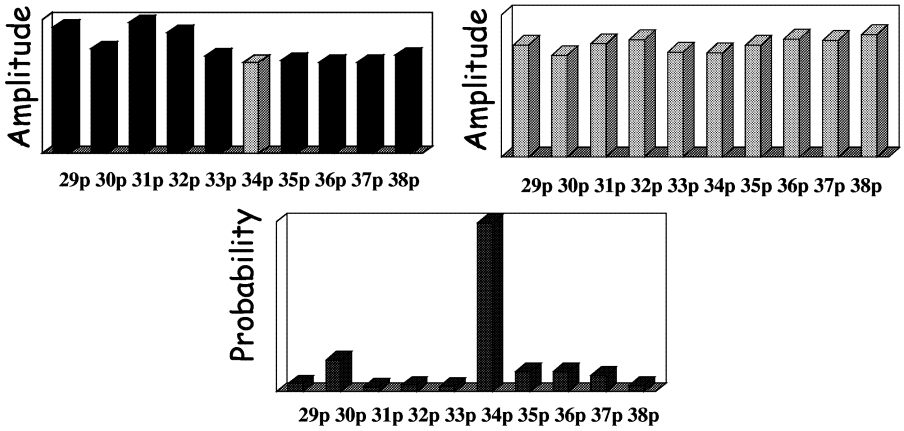


Figure 3. (top left) Bar graph representing a Rydberg data register for the binary number 0000010000. The binary bit is encoded into the phase of the n-state, so that binary 0 states are real and positive (dark phase), while binary 1 states are real and negative (light phase). (Top right) The decoding wave packet has equal amplitudes, and all negative real phases. (Bottom) The superposition of both wave packets amplifies the binary 1 bits, while destructive interference destroys the binary 0 bits.

This is a simple example of a general class of search algorithms that make use of the properties of superposition and quantum interference, which were introduced by Grover.[9] Our particular implementation of a Grover-style search algorithm has some unresolved difficulties if the register is too large. The simple form of the decoding pulse only works in the perturbation theory limit, where almost all of the probability amplitude in the wave packet resides in the "launch" state (7*s* for our work in Cs.) If thre are too many states in the Rydberg wave packet, the launch state will become depleted. The data retrieval is still possible, but now the unitary transformation that amplifies the "1" bits and suppresses the "0" bits must depend on the total pulse energy. In other words, we move into *the strong field regime*.

## Feedback and Learning Control

The techniques of wave packet sculpting are "feed-forward," since the construction and measurement processes make no use make no use of the measured results. A "feedback" loop may be used to adjust the pulse shaper in response to information gathered by wave packet holography. This type of feedback is more properly called "learning," since the readjustment of the apparatus does not occur during a measurement, but rather follows the data acquisition, so that corrections can be made to future experiments.[10] An automated form of learning control on wave packets in Rydberg states can control the shape of the wave packet.[11]

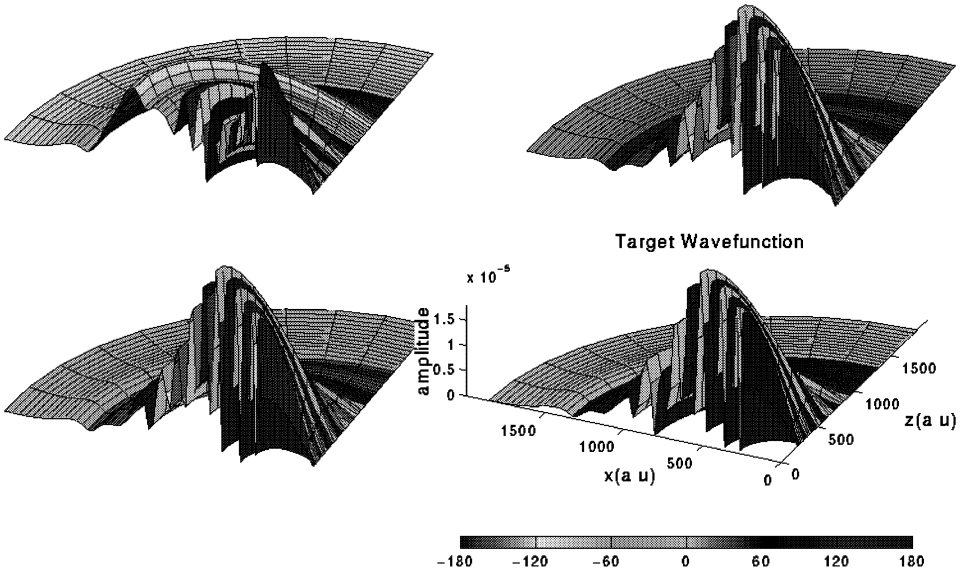


Figure 4. Learning feedback control of a Rydberg wave packet. The desired target wave packet is shown on the lower right. The initial guess produced by the pulse shaper is on the upper left. After two iterations, shown on the upper right and lower left, the experiment converges to the target shape.

This simple feedback control is only effective for systems with a known Hamiltonian and eigenstate spectrum, and for laser interactions in the weak-field limit.

The more general situation is more complicated, but also more useful. Nonlinear optical interactions are used to generate new coherent light sources, and also to probe dynamics. In principle, a strong driving field could be shaped to enhance the desired nonlinear interaction, so that pulse shaping could become a general tool for nonlinear dynamics. Often, the Hamiltonian for any complex quantum system is not known well enough, and with out it one cannot predict likely excitation pathways or derive the optimal pulse shapes. Furthermore, dissipation mechanisms limit the coherence time severely. Learning algorithms for strong field control may aid these problems. In learning control, the experiment runs itself by means of an intelligent feedback loop. It tries various pulse shapes, assesses their success in achieving the desired target excitation, and uses the knowledge gained in this way to improve the pulse shapes on subsequent experiments, all without the intervention of the researcher.

We have been studying learning control in several different nonlinear systems, using a search strategy known as the genetic algorithm (GA).[12] "Genetic" in this case means that the algorithm creates new pulses through a non-local approach based on splicing together traits of successful "parent" laser pulses, rather than by following a fitness gradient function, as in other evolutionary methods.

In our GA implementation, each individual corresponds to a pulse shape, which is encoded as a string of floating point numbers (the individual genome) specifying the phase and amplitude at the various frequency components of the laser pulse. In the first generation, the population consists of sixty individual pulse shapes, chosen at random. We studied quantum dynamics where the target measurement depended on the shape of the driving laser field. The experiment was performed for each pulse shape in turn, and the results assigned a numerical fitness value based on the result. This fitness value determines the chances that a particular pulse shape is selected to reproduce. For example, “roulette wheel” selection, an individual's reproduction probability is proportional to its fitness.

## Selective Excitation of Quantum Modes in Molecular Liquids

Our most successful use of the GA in quantum systems thus far has been in selective excitation of C-H and C-D stretch modes in organic molecules in liquid phase. Control has been demonstrated in methanol, ethanol, benzene, and in mixtures of regular and deuterated benzene ($C_6D_6$). Liquids pose a special challenge to strong field physics, because of rapid relaxation, and also because many nonlinear processes influence the light and make it difficult to define an observable that is related to the desired fitness.

In the liquid phase different experiments were performed, including control of self-phase modulation in carbon tetrachloride and excitation of Raman modes in methanol and benzene. Physical insight is obtained not only through the pulse shapes that the GA finds, but also by how the GA arrived at those solutions.

An example of the kind of control that is possible is shown in the stimulated Raman spectra of methanol excited by GA-shaped pulses. By changing the relative phase between the various spectral components in the pulse, we can selectively excite either the symmetric or asymmetric modes. Further work on state-selective excitation in molecular liquids is underway..

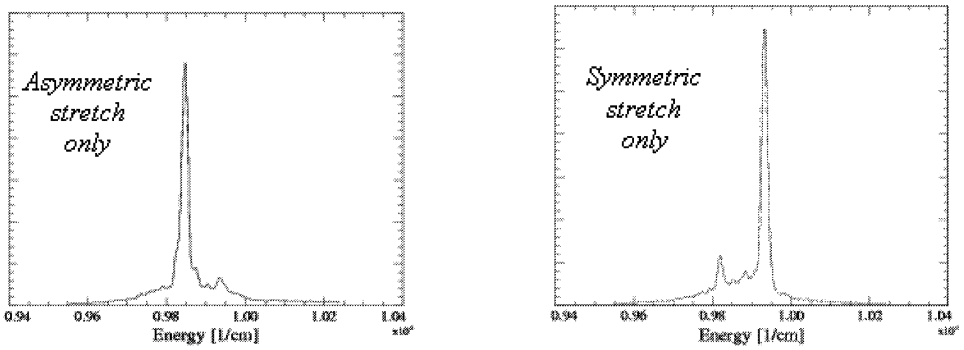


Figure 8. Selective Stokes peaks in the forward scattered light with a methanol sample. The driving laser pulse had a central frequency of 12,700 $cm^{-1}$.

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