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THE KICKED RYDBERG ATOM: NON-LINEAR DYNAMICS AND MANIPULATION OF ATOMIC WAVEFUNCTIONS

by

BRIDGET E. TANNIAN

DOCTOR OF PHILOSOPHY

Houston, Texas
October, 2000
RICE UNIVERSITY

THE KICKED RYDBERG ATOM: NON-LINEAR DYNAMICS AND MANIPULATION OF ATOMIC WAVEFUNCTIONS

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BRIDGET E. TANNIAN

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE DOCTOR OF PHILOSOPHY

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Houston, Texas
October, 2000
ABSTRACT

The Kicked Rydberg Atom: Non-Linear Dynamics and Manipulation of Atomic Wavefunctions

by

Bridget E. Tannian

The response of rubidium Rydberg atoms with principal quantum number $n = 390$ to one or more half-cycle electric field pulses (HCPs) with durations $T_p$ much less than the classical electron orbital period $T_n$ is investigated. High-$n$ atoms subject to a series of HCPs provide a new paradigm, the "kicked atom" for the study of non-linear dynamics in Hamiltonian systems. For certain kick frequencies, dynamical stabilization is observed which corresponds to localization in phase space of the electronic state. The evolution of this localized state is probed with HCPs. A short HCP is used to examine the momentum distribution and a new technique for measuring the spatial distribution of the electron that uses a longer-duration fast-rising is demonstrated. Similar pulses also form the basis of a new approach to the manipulation of atomic $\ell$-state distributions. The experimental data are compared with the results of classical trajectory Monte Carlo simulations. In all cases good agreement between theory and experiment is observed demonstrating that quantum/classical correspondence holds at high-$n$. 
Acknowledgments

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Chapter 1

Introduction

A Rydberg atom is an atom in which one electron is excited to a state of very high principal quantum number $n$. Such atoms have characteristics unlike those of atoms in ground or low-lying excited states. Some of these characteristics are listed in table 1.1 along with their dependence on $n$ and typical numerical values for several $n$. At high $n$, the mean distance between the electron and the atomic core is large, so the gross properties of any species of Rydberg atom resemble those of hydrogen. The orbital period $T_n$ of an electron in a classical Kepler orbit about the core also increases rapidly with $n$. It is of the order of tens of nanoseconds at $n \sim 400$, a time scale that is easily observed on a laboratory oscilloscope. It is therefore straightforward using standard pulse generators to apply unidirectional electric field pulses, termed half-cycle pulses (HCPs), with durations $T_p < < T_n$ to high-$n$ Rydberg atoms. In this limit, the applied field simply delivers an impulsive momentum transfer or "kick" to the excited electron given by

$$
\Delta p = - \int F_{HCP}(t) dt
$$

(1.1)
where $F_{\text{HCP}}$ is the field associated with the HCP. A kick imparts energy to the electron, given by

$$\Delta E = \frac{\Delta \vec{p}^2}{2} + \vec{p} \cdot \Delta \vec{p}$$  \hspace{1cm} (1.2)$$

resulting in coherent excitation of a range of Rydberg states. Sufficiently strong HCPs can deliver enough energy to ionize the atom.

<table>
<thead>
<tr>
<th>Property</th>
<th>classical scaling (a.u.)</th>
<th>$n=1$</th>
<th>$n=30$</th>
<th>$n=400$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean radius</td>
<td>$n^2$</td>
<td>5.3x10^{-9} cm</td>
<td>4.8x10^{-6} cm</td>
<td>8.5x10^{-4} cm</td>
</tr>
<tr>
<td>Orbital Period</td>
<td>$2\pi n^3$</td>
<td>1.5x10^{-16} s</td>
<td>4.1 ps</td>
<td>9.6 ns</td>
</tr>
<tr>
<td>Binding Energy</td>
<td>$\frac{1}{2n^2}$</td>
<td>13.6 eV</td>
<td>15 meV</td>
<td>85 $\mu$eV</td>
</tr>
<tr>
<td>Energy Spacings</td>
<td>$\frac{1}{n^3}$</td>
<td>10.2 eV</td>
<td>1.0 meV</td>
<td>0.43 $\mu$eV</td>
</tr>
<tr>
<td>Classical Field Ionization Threshold</td>
<td>$\frac{1}{16n^3}$</td>
<td>3x10^8 Vcm^{-1}</td>
<td>400 Vcm^{-1}</td>
<td>13 mVcm^{-1}</td>
</tr>
</tbody>
</table>
Measurement of the ionization of Rb(390)p) Rydberg atoms subject to one or more half-cycle pulses provides a tool to investigate the non-linear dynamics of impulsively driven systems and to study the control and manipulation of atomic wavefunctions. Initial measurements using HCPs were undertaken by applying fixed-duration, freely propagating HCPs to Rydberg atoms with low to intermediate $n$. The HCPs were of ~1ps duration and produced by illuminating photoconductive semiconductor switches with output pulses from a femtosecond laser system. These experimental advances stimulated many theoretical investigations. However, the experiments were limited because photoconduction switches cannot produce HCPs of arbitrary shape or trains of closely spaced HCPs. In the present work, measurements have been undertaken at very high $n$, $n = 390$. At such high $n$, required HCP durations are ~1ns, allowing the use of conventional pulse generators to produce HCP pulses and pulse trains. In the present work, the response of atoms to a wide variety of applied fields is investigated, leading to new insights into the dynamical behavior of atomic systems and the control and manipulation of atomic wavefunctions.
Chapter 2  

Background  

In order to understand the effect of an electric field half-cycle pulse on a Rydberg atom in a well-defined initial state, it is necessary first to understand the effect of a static electric field on the motion of the excited electron, the Stark effect. Then the time dependence of the electric field may be considered.

2.1 Hydrogenic Orbits

The hydrogen atom comprises a single electron bound to a proton by the Coulomb potential, $V(r)=-1/r$, where $r$ is the distance between the particles. The classical Hamiltonian is therefore $H = \vec{p}^2 - 1/r$.\(^2\) The potential has rotational symmetry, so the angular momentum of the electron $\vec{L} = \vec{r} \times \vec{p}$ is a constant of the motion. The solutions of $H$ for electron energies $E<0$ are elliptical orbits in a plane normal to $\vec{L}$. The orientation of the major axis of the ellipse is characterized by the Runge-Lenz vector $\vec{M} = \vec{p} \times \vec{r} - \vec{r} / r$, which is also a constant of the motion.

The eccentricity of the orbit is given by\(^3\) $e = \sqrt{1 + 2EL^2}$. Figure 2.1 shows that for low $L$ values, where $e \sim 1$, the orbit is essentially a line between the nucleus and the outer turning point, whereas for large $L$, ($L \sim 1 / \sqrt{2|E|}$) $e \sim 0$, and the orbit becomes a circle.

The position of the electron depends on time as $t = \sqrt{a^3} (\zeta + e \sin \zeta)$.
\[ x = a(\cos \zeta + e), \quad y = a\sqrt{1 - e^2} \sin \zeta \]

where and \(a\) is the semi-major axis, \(a = 1 / 2|E|\). The parameter \(\zeta\), the eccentric anomaly, varies between 0 and \(2\pi\) over one orbital period. The period of revolution depends only on the energy of the electron and is given by

\[ T = 2\pi a^{3/2} = \pi \sqrt{2|E|}. \quad (2.1) \]

The momentum of the electron is

\[ p = \sqrt{2|E|} \left[ \frac{1 - e \cos(\zeta)}{1 + e \cos(\zeta)} \right]^{1/2} \quad (2.2) \]

with

\[ p_x = -\sqrt{2|E|} \frac{\sin \zeta}{(1 - e \cos \zeta)}, \quad p_y = \sqrt{2|E|} \frac{\sqrt{1 - e^2} \cos \zeta}{(1 + e \cos \zeta)}. \quad (2.3) \]

An electron in a low \(L\) orbit moves slowly at the outer turning point and quickly around the nucleus. It spends most of its time on one side of the nucleus. For a high \(L\) orbit, the electron moves approximately in a circular orbit with \(p = p_{\text{rms}} = \sqrt{2|E|} \).

Classical orbits do not correspond one-for-one with quantum-mechanical wavefunctions. However, for \(n \gg 1\), a correspondence can be made between quantum-mechanical wavefunctions and the probability distributions associated with an ensemble of classical orbits with random initial phases.

This correspondence is the basis of classical trajectory Monte Carlo (CTMC) calculations, in which an ensemble of electronic initial conditions are sampled from a classical phase-space probability density which mimics the corresponding quantal position and momentum distributions. Each initial condition in phase space is propagated according to Hamilton's equations of motion.\(^{14}\)
Figure 2.1. (a) Relation of the parameters $a$, $b$, and $e$ to the shape of the orbit. (b) Orbit shapes for various values of scaled angular momentum, $L_0 = L/\sqrt{2|E|}$. 
The large number of states that must be considered when $n \geq 200$ makes quantum mechanical calculations difficult.\textsuperscript{15} However, it is possible to perform quantum mechanical calculations for lower $n$ and scale the results to higher $n$. To limit the number of states used in the calculation, it is desirable to find a basis that spans only the area of Hilbert space in which the electronic state evolves. One possible choice is a finite Sturmian basis set \textsuperscript{16}

$$\Phi_{n,\ell}(r) = \frac{2}{\sqrt{n}} \lambda^{3/2} \left( \frac{(n - \ell - 1)!}{(n + 1)!} \right)^{1/2} (2r\lambda)^{\ell} e^{-\lambda r} r^{2\ell+1} (\lambda r) Y_{\ell}^{m}(\theta, \phi)$$

(2.4)

where the Sturmian parameter $\lambda = 1/n_0$ defines the set. States with $1 \leq n \leq n_{\text{max}}$ are used, which, for $m=0$, corresponds to a number of states $N_{\text{max}} = n_{\text{max}} (n_{\text{max}} + 1) / 2$. The parameters $\lambda$ and $N_{\text{max}}$ are varied to get an accurate result with the smallest possible basis set.

### 2.2 The Stark Effect

The Schrödinger equation for hydrogen in a uniform electric field $\vec{F} = F\hat{z}$ is \textsuperscript{17}

$$(\Delta + 2 / r - 2Fz + 2E) \Psi = 0.$$

(2.5)

The above equation is separable in parabolic coordinates,

$$x = (\xi \eta)^{1/2} \cos \varphi, \quad y = (\xi \eta)^{1/2} \sin \varphi, \quad z = (\xi - \eta).$$

(2.6)

Defining the wave function $\Psi$ as the product $\Psi = (\xi \eta)^{-1/2} V(\xi) U(\eta) e^{i m \phi}$, we obtain

$$\frac{d^2 V}{d\xi^2} + \left( \frac{E}{2} + \frac{\beta_1}{\xi} + \frac{1-m^2}{4\xi^2} - \frac{F}{4} \xi \right) V = 0$$

(2.7)

$$\frac{d^2 U}{d\eta^2} + \left( \frac{E}{2} + \frac{\beta_2}{\eta} + \frac{1-m^2}{4\eta^2} - \frac{F}{4} \eta \right) U = 0$$
where $\beta_1$ and $\beta_2$ are separation constants with $\beta_1+\beta_2=1$.

To second order in $F$, the Stark energy levels calculated using perturbation theory are given by

$$E_{n,n_1,n_2,m} = -\frac{1}{2n^2} + \frac{3}{2} n(n_1 - n_2) F - \frac{n^4}{16} \left[ 17n^2 - 3(n_1 - n_2)^2 + 9m^2 + 19 \right] F^2$$

(2.8)

where $n_1$ and $n_2$ are parabolic quantum numbers with $n = n_1 + n_2 + ml + 1$. Figure 2.3 is a plot of the $H \mid m \rangle = 1$ energy levels from $n=8$ to $n=14$ in electric fields up to $10^5 \text{ Vcm}^{-1}$, strong enough to strip the electron from the atom, resulting in field ionization. Note that the levels shift approximately linearly with $F$ up to field strengths sufficient to ionize the atom. Also, note that the Stark levels of adjacent $n$ manifolds cross, a consequence of the extended dynamical symmetry. The extreme red and blue $m = 0$ states have Stark shifts of approximately $\pm 3n^2 F / 2$. Equating the Stark shift to the $n \rightarrow n+1$ energy spacing gives the Stark crossing field $F = 1 / 3n^5$.

Figure 2.2. Potential energy for a one-electron atom in an electric field $f = 0.02$ a.u. (a) Potential along the $z$ axis; (b) equipotential lines in the $x$-$z$ plane. The point "S" marks the Stark saddle point.
Figure 2.3  Stark structure and field ionization properties of the $|m|=1$ states of the $H$ atom. The zero field manifolds are characterized by the principal quantum number $n$. Quasi-discrete states with lifetime $\tau > 10^{-6}$ s are shown by solid lines, states with lifetimes $5 \times 10^{-10} < \tau < 5 \times 10^{-6}$ by bold lines, and broad field ionizing states with $\tau < 5 \times 10^{-10}$ by broken lines. The saddle point limit $E_c = -2\sqrt{F}$ is shown by a heavy curve. Rapid ionization occurs only for fields $E > E_c$. [from reference 18].
Figure 2.2 shows the electronic potential energy $V(\vec{r}) = -1/r + Fz$ for an atom in an electric field. Classically, field ionization is possible when the electron energy lies above the saddle point located at the local maximum of $V(x=0,y=0,z)$. The position $z_s$ and energy $V_s$ of the saddle point are

$$z_s = -\sqrt{\frac{1}{F}}, \quad V_s = -2\sqrt{F}.$$  \hspace{1cm} (2.9)

Because the potential falls away in the $-z$ direction, there are no true stationary states for an atom in an electric field.\textsuperscript{10} The electron will eventually tunnel through the barrier. For small fields, the tunneling rate is so low that the states are effectively stationary. For strong fields, however, the states ionize so rapidly that they lose their discrete character.

### 2.3 Non-hydrogenic Orbits

The Hamiltonian in equation 2.6 is separable only for a pure Coulomb potential. For atoms other than hydrogen, the atomic core is of finite size and the potential is no longer purely Coulombic. Approximations are made based on the observation that the potential experienced by the outer electron in a neutral atom approaches $-1/r$ for $r$ sufficiently large. For alkali atoms, sufficiently large means larger than the core radius. For large $n$, the electron spends most of its time far from the core ($r \approx n^2$). Thus, the electron wavefunctions can be approximated by Coulombic functions. This observation underlies quantum defect theory, in which the energies of the alkalis can be written

$$E_{nl} = -\frac{1}{2(n - \delta)^2}.$$  \hspace{1cm} (2.10)
where the quantum defect $\delta_i$ is a slowly varying function of $n$. Quantum defects are large for states that penetrate the core and small for states that do not.

### 2.4 Avoided Crossings

Because core penetration breaks the symmetry of the Coulomb orbits when the potential is not a pure Coulomb potential, the Hamiltonian is no longer diagonal in the parabolic basis. The off-diagonal terms correspond to coupling of states of different $n$ and $\ell$. This coupling leads to avoided crossings between Stark states. The energy gap between the states at an avoided crossing depends on the non-Coulombic terms in each state.

$$
\Delta E = 2|\langle n, n_1, n_2, m | V_d(r) | n', n'_1, n'_2, m' \rangle| \tag{2.11}
$$

where $V_d$ is the difference between the real potential and a pure Coulomb potential.\textsuperscript{18}

For low $m$, $|m| < n$, the energy separation at avoided crossings between Stark states from neighboring $n$ manifolds scales approximately as $1/n^4$. States of low $\ell$ have a lot of core interaction and therefore exhibit larger energy gaps at avoided crossings than do states of high $\ell$.\textsuperscript{19}

### 2.5 Adiabatic and Diabatic Ionization

The evolution of states in a time-dependent electric field is strongly dependent on the slew rate of the field. If the field increases slowly, the electronic state will be able to respond to changes in the field and follow an adiabatic path through an avoided crossing.
If the field increases rapidly, the wave function will not be able to change in response to the field and the character of the state will stay the same. The atom will "jump" over the avoided crossing diabatically. Adiabatic and diabatic transitions are illustrated schematically in figure 2.4.

The probability of traversing an avoided crossing diabatically is given by the Landau-Zener formula

\[ P = \exp \left[ -\frac{\pi |V|^2}{\hbar (dW/dt)} \right] \]  

(2.12)

where \( V \) is the matrix element for the avoided crossing \( (2V \) is the separation) and \( dW/dt \) is the change in level separation as the crossing is approached, \( \frac{dW}{dt} = \left( \frac{dW}{dF} \right) \left( \frac{dF}{dt} \right) \). The probability of a diabatic passage across the avoided crossing increases with increasing slew rate.

![Figure 2.4. Adiabatic or diabatic passage of state B through an avoided crossing with state A.](image-url)
2.6 Classical Scaling

The classical scaling of the quantities in table 1.1 is derived from the fact that the classical equations of motion are invariant under the length, momentum, time, and field transformation $r = \alpha^2 r_\alpha$, $p = \alpha^{-1} p_\alpha$, $t = \alpha^3 t_\alpha$, $F = \alpha^4 F_\alpha$, where $\alpha$ is a scaling parameter. In the scaled variables, the atomic Hamiltonian $H_\alpha(t_\alpha) = \alpha^2 H(t)$ is independent of $\alpha$. The Hamiltonian remains independent of $\alpha$ when an external electric field pulse is applied if the time-dependent field is of the form $F_g(t/T_p)$, where $T_p$ is the duration of the field pulse. A convenient choice of scaling parameter for time-dependent problems is $\alpha = n_i$ (the initial value of $n$) so that the duration of the pulse is referred to the orbital period $T_o$.

2.7 Chaos in Hamiltonian Systems

A Hamiltonian system of dimension $2M$ is integrable if there exist $M-1$ functions such that

$$\frac{d}{dt} F(x(t)) = \sum_{i=1}^{M} \left( \frac{\partial F}{\partial q_i} \frac{\partial H}{\partial p_i} - \frac{\partial F}{\partial p_i} \frac{\partial H}{\partial q_i} \right) = \{F, H\}$$

and

$$\{F_i, F_j\} = 0$$

Each orbit of an integrable system is contained on a $M$-dimensional torus, resulting from the multiple intersection of the $M$ surfaces $F_j(x) = \text{const.}$, each with $(2M - 1)$ dimensions. An integrable system is not chaotic. However, of the many mechanical systems of physical significance, few are known to be integrable. Non-integrable systems may
exhibit regular behavior, fully chaotic behavior, or a mixture in which the motion is regular or chaotic for different initial conditions.

The Poincaré surface of section provides a means for visualizing complicated dynamics. Poincaré suggested reducing the dimensionality of the phase space flow of a dynamical system by intersecting the energy surface with another suitable chosen subspace of phase space such that the intersection of the two manifolds is one dimension lower than the dimensionality of the energy surface.\(^{21}\) The resulting lower-dimensional space is called the surface of section. To construct a Poincaré plot, a convenient surface in phase space and an initial condition for the trajectory are chosen. Each time the trajectory passes through the surface in one direction, a point is plotted at the point of intersection. Regular (periodic or quasiperiodic) motion produces a smooth curve or a dot that is a cross section of the torus to which the motion is constrained. Chaotic motion generates a seemingly random set of points filling the space. Figure 2.5 shows a Poincaré surface of section for an unperturbed 1D atom.\(^{22}\) (For 1D motion, the Poincaré surface of section is simply a stroboscopic picture of the motion.) The lines correspond to tori for different atomic states.

The transition from an integrable system to a non-integrable system through a small perturbation is the subject of the Kolmogorov-Arnold-Moser (KAM) theorem, which states that for an integrable system with a small perturbation

\[
H = H_0 + \epsilon H_1
\]  

(2.15)

almost all tori are preserved.\(^{21}\) That is, almost all solutions in the perturbed system are confined to invariant tori that are slightly deformed from the tori of the unperturbed
system. These surviving tori are referred to as KAM tori. For mixed systems, KAM tori appear in Poincaré plots as islands in a chaotic sea.

When the perturbation grows large enough that an island is destroyed, an invariant cantor set known as a cantorus remains. Cantori impede the flux of phase points in the chaotic regions of mixed systems and thus produce partial confinement. For systems with more than two degrees of freedom, invariant tori and cantori do not have sufficiently high dimension to confine the motion in the chaotic regions of most mixed systems. Chaotic motion around the tori, known as Arnol'd diffusion, can then take place.

Figure 2.5. Poincare map of a 1-D atom. The x-axis represents scaled coordinate and the y-axis scaled momentum.
In the present work, high-\(n\) Rydberg atoms are created by photoexciting rubidium atoms in a collimated beam by a frequency doubled tunable ring dye laser. Excitation occurs near the center of an interaction region, shown in figure 3.2. The interaction region is defined by three pairs of planar copper electrodes that are independently biasable in order to minimize stray electric fields. Experiments are conducted in a pulsed mode. Following each laser pulse, the Rydberg atoms are exposed to half-cycle pulses. Measurements in which no HCPs are applied are interspersed at routine intervals during the data acquisition. The Rydberg atom survival probability is obtained by comparing the Rydberg atom signals observed with and without HCPs present.

3.1 Vacuum System

The vacuum system comprises two chambers separated by a 0.5mm aperture. One contains the oven used to produce the Rb beam, the other the main experimental region. The chamber containing the rubidium oven is pumped by a 4" diffusion pump. The main chamber contains the interaction region where Rydberg atoms are created and ionized. It is pumped by a 6" diffusion pump. The pressure in the main chamber is measured using a Bayard-Alpert type ionization gauge and is typically \(~1 \times 10^{-7}\) torr. The laser beam enters and exits the main chamber through quartz windows set at Brewster's angle.
3.2 Rubidium Beam

The rubidium oven is an enclosed stainless steel reservoir with a 0.5mm orifice. The reservoir is heated to 255°C and the exit hole to 280°C by resistive cartridge heaters. The oven temperature is monitored by thermocouples and controlled by microprocessor temperature controllers. The exit orifice is set at a higher temperature to prevent rubidium from condensing there and blocking it. The rubidium beam enters the main chamber through a 0.5mm collimating aperture, resulting in a beam divergence of ≤1°. The beam density is estimated to be ~10⁹ cm⁻³ at the center of the interaction region.²⁴ The average velocity of the rubidium atoms is ~300 m/s. The beam contains the two rubidium isotopes in approximately their natural abundance; 72% $^{85}$Rb, 28% $^{87}$Rb.

3.3 Laser System

The wavelength needed to excite $n=390$ rubidium Rydberg atoms by single-photon excitation is 2968.24 Å. This is provided by an intracavity frequency doubled ring dye laser, a Coherent model CR-699-21 with a model 7500-06 doubling package, shown schematically in figure 3.1. This is pumped by a Coherent Innova 400 Argon Ion laser. 9.7 W of 514.5 nm light from the argon ion laser is focused onto a stream of Rhodamine 6G Chloride dye in the ring laser. The dye has a continuous fluorescence spectrum that allows the laser to be tuned over a range of wavelengths. Single frequency operation is obtained through the use of a three-plate birefringent filter, a thin etalon, and a piezoelectric-driven thick etalon. The birefringent filter has a passive bandwidth of about 380 GHz, and the thin etalon has a free spectral range of 200 GHz, which confines lasing to a single longitudinal mode.²⁵ The thick etalon permits the longitudinal mode to be
scanned. The radiation circulating in the cavity passes through a non-linear potassium dihydrogen phosphate (KDP) crystal that converts a small fraction of it into UV light by second harmonic generation, producing 1-3 mW of UV output.

Figure 3.1. Schematic diagram of the Coherent 699 in dye laser.

The laser frequency is stabilized using a temperature controlled Fabry-Perot etalon as a reference. Deviations in frequency from the lock point are corrected by changing the optical length of the laser cavity. To correct fast deviations, the tweeter mirror is mounted on a PZT. Slow drifts are corrected by rotating the galvo-driven quartz plate about Brewster's angle.

The reference cavity drifts enough in temperature to shift the lock point by ~40 MHz/hr in the fundamental. To achieve the frequency stability necessary for the present experiments, the laser is locked to a frequency stabilized Melles Griot helium-neon laser using another temperature controlled Fabry-Perot etalon. The HeNe and dye laser beams are superposed and directed through the etalon. The etalon mirror spacing is
scanned at 50 Hz, producing two series of transmission peaks, one for each wavelength. Drifts in the dye laser frequency result in changes in the separation between adjacent HeNe and dye peaks. These changes are used to generate error signals to restore the laser to the lock point. Using this system, the overall frequency stability of the dye laser is reduced to ±1 MHz/day in the fundamental. By changing the lock point, the laser can be scanned over a 500 MHz range.

The laser wavelength is monitored using a scanning Michelson interferometer. The wavemeter compares the number of fringes measured with the fundamental output of the dye laser to the number measured with a polarization stabilized HeNe directed through the same apparatus. The accuracy of the wavemeter is ±1 GHz at the operating frequency.

The UV output of the dye laser is chopped into 4μs pulses using an acousto-optic modulator (AOM). In the AOM, a PZT generates an RF acoustic traveling wave in a piece of fused quartz. The periodic variation of the refractive index of the quartz creates a diffraction grating. The sound wave is pulsed and the first order of the diffracted UV light is directed into the interaction region.

3.4 Interaction Region

The interaction region is in the center of the main chamber, where the laser is focused onto the rubidium beam. It is surrounded by three pairs of copper electrodes, each 10 cm x 10 cm (see fig. 3.2). The use of large electrodes far from the experimental volume minimizes the variation of the electric field over the experimental volume and the effect
of patch fields associated with non-uniformities in the electrode surfaces. Electric field HCPs are produced in the interaction region by applying voltage pulses to a circular copper disk, 5 cm in diameter that is mounted on the end of a section of semi-rigid coaxial cable and positioned 2.5 cm below the upper rectangular electrode. To prevent pulse reflection, the electrode is grounded with a 50 Ω resistor. To reduce RF pickup on the other electrodes, the top plate is grounded and RF-decoupling capacitors are connected between the side plates and ground.

DC electric fields in the interaction region are reduced to ≤ 50 μV/cm by the technique described in reference 27. Magnetic fields are reduced to ~20 mG by a μ-metal shield that surrounds the interaction region.
The location of the HCP electrode in the copper box minimizes the stray capacitance of the electrode, allowing fast pulse rise times to be obtained. Also, the symmetry of the interaction region is reduced, limiting the excitation of cavity resonances. One problem when using very short HCPs is that of relating the electric field experienced by an atom in the center of the interaction region to the voltage pulse applied to the HCP electrode. For sufficiently long pulses, the electric field can be determined accurately by solving Laplace's equation with appropriate boundary conditions. Such calculations were performed to ensure that the DC electric field near the center of the interaction region did not vary by more than 50 μV/cm within the experimental volume. For the fastest pulses, however, the pulse duration is only twice the transit time of an electromagnetic pulse across the interaction region. Calculation of the electric field therefore requires solution of Maxwell's equations. Rather than attempt this, the ionization of high-\(n\) Rydberg atoms was used to calibrate the field. The probability for ionization of Rb(390p) atoms by a single HCP was measured as a function of HCP amplitude and width. Previous work \textsuperscript{28} using higher-\(n\) atoms and longer-duration HCPs (\(\sim\)2ns) has shown that CTMC simulations accurately predict absolute survival probabilities. Thus, by comparing theory and experiment, it is possible to determine the impulse delivered by a given HCP. The calibration data for pulses from the HP8131A pulser are shown in figure 3.3.

Rydberg atoms remaining in the interaction region after HCP application are ionized by the electric field generated by a slow voltage ramp applied to the bottom plate. Electrons resulting from ionization are accelerated by the extraction ramp through a mesh-covered hole in the bottom plate to a Johnson Laboratory MM-1 electron multiplier for detection.
Figure 3.3. Calibration of electric field pulses generated by applying voltage pulses to the HCP electrode. The filled circles represent the results of CTMC calculations of the impulses necessary to ionize 10% of the high-n atoms using gaussian-shaped pulses. The open circles are the experimentally measured impulses assuming that the amplitude of the applied field is equal to that calculated for a DC field.

3.5 Half-Cycle Pulses

The half-cycle pulses (HCPs) are produced using (variously) a Hewlett Packard 8082A, a Hewlett Packard 8131A, and an Avtech AVI-V pulse generator. The HP pulsers can generate pulses with amplitudes up to ~5 V. The 8131A can produce pulses with widths as short as 400ps FWHM and the 8082A as short as 2ns FWHM. The amplitudes of the pulses are adjusted using broadband (DC - 2 GHz) 50 Ω attenuators placed in series with the pulse generator output. In order to supply both a DC bias and a
HCP to the HCP electrode, the pulses pass through an Avtech AVX-TB bias tee before going to the HCP electrode.

The pulses are measured directly at the HCP electrode using a Tektronics P6156 DC-3.5 GHz voltage probe and a Tektronics 7603 sampling oscilloscope with an S-2 type sampling head. Spatial variations in the field produced by the HCP electrode and uncertainties inherent in the measurement of HCP amplitude introduce an uncertainty of \( \leq \pm 10\% \) in the applied fields.
Chapter 4
Experimental Techniques

4.1 Experimental Methods

Rydberg atoms are created by photoexciting rubidium using a UV laser pulse. The laser output is chopped into a train of pulses and measurements are conducted in pulsed mode. The probability that a Rydberg atom is formed during any laser pulse is small, $\leq 0.05$, and data are accumulated over many cycles. Following each laser pulse, the Rydberg atom is exposed to one or more half-cycle pulses (HCPs), which are generated by applying voltage pulses to a small circular copper disc hanging below the top electrode. Measurements in which no HCPs are applied are interspersed at routine intervals during the data acquisition. The Rydberg atom survival probability is obtained by comparing the Rydberg atom signals observed with and without HCPs present.

Data are accumulated under computer control. The master pulse generator produces pulses at $\sim 25$ kHz to begin the timing sequence shown in figure 4.1. One output triggers the acousto-optic modulator which generates a 4$\mu$s laser pulse for exciting a Rydberg atom. Another output triggers the computer-controlled gate (CCG) which, when enabled by the computer, triggers a SRS DG535 delay generator. The DG535 controls the delay or the width of HCPs by changing the delay between its output pulses. A third pulse from the master pulser triggers the Evans 4145 programmable delay module, which triggers the SFI ramp and the TAC at a selected delay time. Electrons resulting from field ionization are pulled out of the interaction region by the field ramp
and detected by the MM-1 electron multiplier. Figure 4.2 shows the setup of the data acquisition system.

4.2 Selective Field Ionization (SFI)

Selective field ionization (SFI) is a technique for detection of Rydberg atoms in which a steadily rising electric field is applied across the interaction region to ionize any Rydberg atoms present. Because atoms in different Rydberg states ionize in different fields, measurement of the time dependence of the ionization signal gives an indication of the initial states of the Rydberg atoms. Thus, SFI is useful both to detect Rydberg atoms and to determine their excited state distribution.

The physics of ionization by ramped electric fields is treated in more detail elsewhere.\textsuperscript{18,29,30} In small fields, less than the field at which states from adjacent Stark manifolds start to cross, $\sim 1/3n^5$, the parabolic $nn_1n_2m$ states evolve adiabatically in the field. As the field becomes greater than $\sim 1/3n^5$, avoided crossings occur with states of the same $m$ and different $n$. At $n\sim 400$, the frequency separations between adjacent Stark states at the avoided crossings are much less than the rate at which the applied ramp rises, so the avoided crossings are traversed diabatically. As in hydrogen, ionization begins at $F\sim 1/16n^4$. As the ramp rises, atoms with higher values of $n$ ionize before atoms with lower values of $n$. Thus the time of arrival of the electrons at the detector can give information on the initial distribution of excited states.

The free electrons produced by HCP-induced ionization will also be extracted by the field ramp and can contaminate the SFI spectrum. Thus there must be a delay between
Figure 4.1. Data acquisition timing sequence. The pulse amplitudes are not to scale. The pulses represented by broken lines are gated off in alternate sets of experimental cycles.
Figure 4.2. Block diagram of the data acquisition system.

the HCP and the start of the SFI ramp sufficient to allow these electrons to drift out of the collection volume. The residence times for free electrons have been measured to be $\leq 5$ $\mu s$. For the present work, delay times of 7-10 $\mu s$ were used.

4.3 High-$n$ Spectroscopy

The experiments in this thesis were performed using the $390p$ state of $^{85}$Rb. The
excitation wavelength for this state, given by

$$\frac{hc}{\lambda} = E(n,p) - E(5,s,F),$$

(4.1)

is 2868.24 angstroms. The wavemeter is not sufficiently accurate to determine which Rydberg state is being excited by the laser, so the laser is scanned over a range of frequencies. The resulting Rydberg atom spectrum is compared to a calculated spectrum and the laser frequency is adjusted appropriately.

Figure 4.3.a shows a scan about $n\sim390$. Note that there are four Rydberg series contributing to the signal. The two isotopes of rubidium each have two ground-state hyperfine levels. The width of the lines, $\sim10$MHz, is mostly due to Doppler broadening resulting from the divergence of the rubidium beam.

The calculated rubidium spectrum appears in figure 4.3.b. The energy levels of high-$n$ rubidium can be approximated as

$$E(n,\ell) = -\frac{R}{[n - \delta(n,\ell)]^2}$$

(4.2)

where $R$ is the Rydberg constant and $\delta(n,\ell)$ is the quantum defect. $\delta$ was evaluated using a modified Rydberg-Ritz formula

$$\delta(n,\ell) = a_0 + \frac{a_1}{(n-a_0)^2} + \frac{a_2}{(n-a_0)^4} + \frac{a_3}{(n-a_0)^6} + \cdots$$

(4.3)

with constants found in the literature.$^{31}$
Figure 4.3. a. Rydberg atom signal observed as the laser is scanned over a 800MHz interval. b. Calculated high-\(n\) spectrum of Rb. The x axis is the detuning from the fundamental laser frequency required to excite the 85Rb F=2 390p state in MHz.
Chapter 5

Experimental Results and Discussion

5.1 Manipulation of the Atomic $\ell$-state Distribution

Photoexcitation of atoms from the ground state permits highly selective excitation of a single, well-defined high-$n$ state. However, only states with small values of the orbital angular momentum quantum number $\ell$ are directly accessible. Previously proposed techniques for creating high-$\ell$ states are based on adiabatic manipulation of the Rydberg electron wavefunction. The time scale that defines the adiabatic regime is determined by the Larmor period of the Rydberg electron. In the present work, a new approach to the generation of high-$\ell$ states has been demonstrated based on purely diabatic transitions.

In this experiment, Rb(390np) Rydberg atoms are subject to a large-amplitude rectangular electric field pulse whose rise and fall times are short compared to the electron orbital period. Classically, a simple quasiperiodic evolution of the wavepacket is expected corresponding to the precession of the electron orbit in the applied field. This evolution results in periodic changes in the electron orbital angular momentum at the Stark frequency $\omega_s = 3nF$, where $F$ is the applied field. For $n=390$ and $F=10$ mV/cm, $T_s \sim 67$ ns. Figure 5.1.a shows the calculated evolution of the angular momentum $L$ of the Rydberg electron after application of a 10 mV/cm field step. At early times, the $L$
Figure 5.1. (a) Time evolution of the distribution of electron orbital angular momentum $L$ following diabatic application of a 10 mV/cm electric field at $t = 0$ to rubidium atoms initially excited to the 390p state. (b) Corresponding time dependence of the expectation value of $L$ and $L_x$. The times labeled A-D correspond to the times at which the rectangular field is turned off in figures 5.3 and 5.4.
distribution is tightly peaked at small values. As the electron orbit precesses, the $L$
distribution remains narrow, but moves to higher $L$. Figure 5.1.b shows the time
development of $<L>$ and $<L_z>$. The expectation value of the orbital angular momentum
peaks at $<L> \sim 310$ after one half of a Stark period. The electron angular momentum
then begins to decrease until it becomes very small again after a full Stark period. The
periodic variations in $L$ continue as long as the DC field is applied. When the field is
suddenly turned off, the $L$ distribution remains frozen where it was at the time of the turn-
on. Thus, by varying the length of the rectangular electric field pulse, it is possible to
manipulate the final $L$ distribution.

Quantum mechanically, rapid application of an electric field projects the initial
state $|i> = |n_f, \ell_i, m_i>$ into the Stark manifold, forming a coherent superposition of Stark
states that can be written, in the sudden approximation, as

$$|\Psi(t)\rangle \equiv \sum_\alpha e^{-iE_\alpha t} \langle \alpha | i \rangle |\alpha\rangle$$

where $E_\alpha$ and $|\alpha\rangle$ are the eigenenergies and eigenvectors in the field. As described in
chapter 2, the frequency separations between adjacent levels in a given Stark manifold
are given by $\Delta \epsilon = \omega_s = 3nF$. The amplitudes of the rectangular pulses, $\sim 10$ mV/cm, are
such that states from several neighboring Stark manifolds overlap. (The Stark crossing
field is $1/3n^5 = 0.2$ mV/cm.) It has been shown\textsuperscript{34} that the Stark states with significant
occupation probability are spaced such that the resulting frequency spectrum is strongly
peaked at the Larmor frequency. Diabatic projection into the Stark manifold causes this
hydrogenic behavior. Thus, we expect the period of evolution of $\langle L \rangle$ to match that predicted by the classical simulations.

The final L distributions after applying rectangular electric field pulses are probed using a half-cycle pulse with a width of $\sim 2$ ns. This HCP delivered an impulsive momentum transfer, or "kick", $\Delta p_z$ to the excited electron, where $z$ is the direction of the electric field generated by the HCP. The resulting change in energy is given by equation 1.2. For a given impulse $\Delta p_z$, the probability that the Rydberg atom will be ionized depends on the $z$ component of the electron momentum at the time of the kick. The variation in the $z$ component of the electron momentum after the rectangular pulse is turned off depends on the final $L$ distribution. For small values of $\langle L \rangle$, the $z$ component of the angular momentum will not vary much with time. Thus, the Rydberg atom survival probability will not be very sensitive to the delay between the end of the rectangular pulse and the application of the probe pulse varies. For large values of $\langle L \rangle$, the survival probability varies much more with pulse delay.

The setup used to produce a relative delay between the rectangular pulse and the HCP probe appears in figure 5.2. The rise time of the rectangular pulse is $\sim 3$ ns. Its width can be varied from zero to several $\mu$s. The field amplitude decreases by $\sim 5\%$ over 200 ns, the longest pulse used, because the voltage pulse that produces the field is AC coupled through a bias T to ensure a stable DC baseline over the course of the measurement.

Figure 5.3 shows the measured Rydberg atom survival probabilities as a function of delay between the end of the rectangular pulse and the HCP. These span one Stark
Figure 5.2. Setup to produce a relative delay between the rectangular pulse and the HCP probe pulse.

...period and correspond to the times labeled A-D in figure 5.1.b. The observed survival probabilities depend markedly on the position in the Stark cycle at which the electric field is turned off.

Pronounced periodic oscillations in the survival probability are observed in each data set. These oscillations occur because the diabatic turn on / turn off of the rectangular field pulse creates a coherent superposition of final states that encompass a small range \( \Delta n \) of \( n \) values centered on the initial laser-excited state. The frequency of these quantum beats is governed by the spacing between the various \( n \) levels. For a difference \( \delta n \) in \( n \), this spacing is given to second order in \( \delta n / n \) by

\[
E_{n+\delta n} - E_n = \delta n \omega_0 \left[ 1 - \frac{3\delta n}{2n} + 2\left( \frac{\delta n}{n} \right)^2 \right] \tag{5.2}
\]

where \( |\delta n| \leq \Delta n/2 \ll n \). To first order, the frequency spectrum locally resembles that of a harmonic oscillator where \( f_n = 1/T_n \) is the classical orbital electron frequency. For \( n \sim 390 \), the predicted quantum beat frequency is \( \sim 110 \, \text{MHz} \), which agrees well with that observed. The beats damp because beyond the first order the Rydberg levels are not...
equispaced in energy. CTMC calculations suggest population of states spanning a range
\( \Delta n \sim 8 \), leading to a predicted dephasing time \( t_D \sim (4n/3\Delta n^2)T_n \sim 72\text{ns} \).

Figure 5.4 shows measured and calculated survival probabilities for the time A and C in figure 5.1.b which correspond to extremes in the final \( L \) distribution. The calculations use the measured pulse profiles and contain no adjustable parameters. They also take into account the small residual field that remains in the interaction region following the application of the rectangular pulse, which ionizes atoms with \( n > 800 \). In the calculations, it is assumed that \( p \) states with a statistical distribution of \( m \) are formed initially. This corresponds to a subset of a microcanonical ensemble with the classical angular momentum restricted to \( [\ell, \ell+1] \) with \( \ell=1 \). Each initial condition in phase space is propagated according to Hamilton's equation of motion taking into account the experimentally determined time dependence of the HCP field. Ionization occurs when the final energy of the electron is greater than the ionization threshold energy, which is zero when there is no stray electric field. The interaction between the electron and the \( \text{Rb}^+ \) core is represented by a model potential that yields accurate quantum defects and satisfies correct boundary conditions at small and large distances. \(^6\)

The initial amplitudes of the observed quantum beats are well reproduced by the CTMC calculations. The observed beats damp more rapidly than predicted, which might be related to field inhomogeneities in the interaction region. Product states with the largest values of \( \ell \) yield the largest quantum beat amplitudes, and these amplitudes provide a useful indicator of high-\( \ell \) state production. High-\( \ell \) states produced by this method may be used as initial states in other experiments.
Figure 5.3. Rydberg atom survival probabilities as a function of time delay between the end of the rectangular pulse and application of the probe HCP for rectangular field pulses of 10mV/cm and durations corresponding to the times labeled A-D in figure 5.1.b.
Figure 5.4. Comparison of the measured (*) and calculated (---) Rydberg atom survival probabilities following application of 10mV/cm rectangular pulses with durations corresponding to the times A and C indicated in figure 5.1.b. The calculations assume that, following HCP application, atoms in states with $n > 800$ are ionized by the residual field.
5.2 Response to Trains of Impulses

Studies of the behavior of impulsively driven, or "kicked," systems in which the duration of each impulse is short compared to the period of the unperturbed system can provide valuable insights into nonlinear dynamics in Hamiltonian systems. Impulsively driven systems, which include the kicked rotor and the kicked hydrogen atom, are straightforward to model numerically because their time evolution can be reduced to a series of discrete maps between adjacent kicks. Although systems subject to sinusoidal perturbations have been widely studied, for example, hydrogen Rydberg atoms in a microwave field \(^{35}\), experimental realizations of kicked systems are few. Recently, atoms cooled in a standing light wave have provided a practical example of the one-dimensional standard map and have demonstrated the existence of Anderson localization in the translational motion of a single atom.\(^{36}\) Previously in this lab, the kicked atom has been realized by exposing high-\(n\) potassium Rydberg atoms to HCPs whose duration is short compared to the classical electron orbital period. However, in these studies, the pulse trains were limited to scaled frequencies \(\nu_o \equiv \nu_p / \nu_n \leq 2\) and to scaled pulse widths \(T_o \equiv T_p / T_n \geq 0.2\), barely in the impulsive limit.

In the present work, measurements are extended to higher scaled frequencies, \(\nu_o \sim 4\), and to shorter HCP widths, \(T_p \sim 500\)ps FWHM. The shorter pulse width \((T_p / T_n - 0.05)\) provides better access to the (near) impulsive limit. New measurements also include trains of HCPs that alternate in sign, i.e., that provide kicks that alternate in direction. Typical segments of the HCP trains appear in figure 5.5.
Survival probabilities measured following application of trains of 20 and 50 equispaced, unidirectional HCPs are presented in figures 5.6.a and 5.6.b respectively as a function of the pulse repetition frequency \( v_p \) for several HCP amplitudes. As might be expected, significantly larger HCP amplitudes are necessary to obtain a given fractional ionization when using 20 HCPs than when using 50 HCPs. The data show that the survival probability depends markedly on the pulse repetition frequency and a pronounced maximum is evident at scaled frequencies \( v_o \approx 1.3 \). The survival probability at the maximum is surprisingly large. A HCP amplitude of 180 mV/cm corresponds to a scaled momentum transfer \( \Delta p \equiv \Delta p/p_n = n\Delta p \approx 0.33 \), suggesting 50 HCPs should be more than sufficient to ionize essentially all the atoms present.

The survival probabilities observed after application of 20 and 50 HCPs that alternate in sign are shown in figures 5.7.a and 5.7.b respectively as a function of pulse repetition frequency. The pulse repetition frequency is taken to be \( 1/T_p \) where \( T_p \) is the
time between adjacent HCPs. The survival probability again depends strongly on \( \nu_p \), but, in contrast to the data for unidirectional HCPs, no broad peak in survival probability is evident. Rather, the survival probabilities generally increase with increasing pulse repetition frequency. A small local maximum in survival probability is observed, however, for scaled frequencies \( \nu_o \sim 1 \).

Figures 5.6 and 5.7 include the results of 3D CTMC simulations, which agree well with the data without recourse to any adjustable parameters. In the case of alternating HCPs, however, the calculated survival probability was found to be quite sensitive to the presence of small residual fields. No similar sensitivity exists for unidirectional HCPs because the time-averaged electric field associated with the train of pulses \( F_{av} = \nu_p \Delta p \) is much larger than any residual field. The time-averaged field for alternating pulses is \( F_{av} = 0 \) and, therefore, stray fields play a comparatively larger role. The CTMC results in figure 5.7 assume the presence of a small residual field sufficient to ionize atoms with \( n \geq 1200 \). The size of the calculated peak predicted at scaled frequencies \( \nu_o \sim 1 \) is somewhat larger than that measured. In addition, the calculations exhibit a local maximum at \( \nu_o \sim 0.5 \) that is not visible in the measurements. Given the sensitivity to stray fields, these differences could be the result of small field inhomogeneities in the experimental volume.

The origin of the features present in the data was examined by considering the behavior of the system when it is subject to a large number of \( \delta \)-function impulses. CTMC simulations indicate that survival probabilities calculated for \( H(390p) \) using this
Figure 5.6. (a) Rb(390p) Rydberg atom survival probability following application of twenty unidirectional HCPs with peak fields of ■ 200, _ 225, ◦ 250, ▲ 275, and ▼ 300 mV/cm as a function of pulse repetition frequency, expressed in MHz and scaled units $\nu_o = \nu_p / \nu_n$. The figure includes the results of CTMC simulations that use the measured HCP profile and pulse amplitudes of (---) 200 and (---) 275 mV/cm. (b) Survival probability following application of 50 unidirectional HCPs with peak fields of ■ 120, _ 150, and ◦ 180 mV/cm. The results of CTMC simulations for pulse amplitudes of (---) 120 and (---) 180 mV/cm are also shown.
Figure 5.7. (a) Rb(390p) Rydberg atom survival probability following application of twenty alternating HCPs with peak fields of ■ 200, □ 225, ○ 250, ▲ 275, and ▼ 300 mV/cm as a function of pulse repetition frequency. The lines show the results of CTMC simulations that use the measured HCP profile and pulse amplitudes of (—) 200 and (—–) 275 mV/cm. (b) Survival probability following application of 50 alternating HCPs with peak fields of ■ 120, □ 150, and ♦ 180 mV/cm. The results of CTMC simulations for pulse amplitudes of (—) 120 and (—–) 180 mV/cm are also shown. The CTMC simulations assume the presence of a field sufficient to ionize atoms with \( n > 1200 \).
simplified model and for Rb(390p) using finite width pulses display the same general structure, demonstrating that use of the simplified model is reasonable. The broad maximum in the survival probability for unidirectional pulses comprises a series of overlapping peaks whose origin can be traced by analyzing Poincaré surfaces of section. Each of the peaks in figure 5.6 corresponds to a regular island in phase space associated with dynamical stabilization. The survival probability after many kicks is given by the fraction of the initial phase-space points that are contained within a given island. This is illustrated in figure 5.8, which shows Poincaré maps for different pulse repetition frequencies together with the fixed initial torus. For repetition frequencies \( v_o = 0.616, 1.07, \) and 2.12, the initial torus overlaps with different stable islands and, therefore, stabilization is obtained by trapping the phase-space trajectories within a given island. For \( v_o = 0.701, 1.978, \) and 5.554, the initial torus lies in the chaotic sea and the atom becomes fully ionized. A further illustration of dynamical stabilization appears in figure 5.9.a, which shows the calculated survival probability as a function of the number \( N \) of kicks for several values of scaled frequency. When the initial state lies within a stable island, the survival probability, after initially decreasing, becomes steady, even for \( >10^6 \) kicks. For other values of frequency, the survival probability decreases monotonically with \( N \), a characteristic of chaotic ionization.

Additional evidence for dynamical stabilization and chaotic ionization is contained in the distribution of excited states following application of the HCP train. If dynamical stabilization occurs, the distribution of excitation energies of the surviving bound states should be sharply peaked near the initial energy. This behavior is confirmed
by the SFI spectra shown in figure 5.10. In the absence of HCPs, the SFI spectrum comprises a single relatively narrow peak that corresponds to ionization of parent 390p atoms. The SFI profile observed following application of 50 HCPs with a scaled repetition frequency $v_o \sim 1.3$ (for which the survival probability is near maximum) and an amplitude sufficient to ionize $\sim 90\%$ of the parent atoms is broader than that for the parent. This indicates that HCP application populates a range of final $n$ states. The majority of the atoms, however, ionize at field strengths comparable to those of the parent atoms, demonstrating that the $n$ distribution of the surviving atoms is peaked at a value of $n$ similar to that of the parent atoms, as is expected if dynamical stabilization is occurring. The SFI profile for a scaled repetition frequency $v_o \sim 0.5$, where dynamical stabilization is not expected, contains no pronounced feature at field strengths near those characteristic of parent-state ionization. Rather, the SFI spectrum is dominated by the ionization of very-high-$n$ atoms, which is consistent with chaotic migration from the initial state to highly excited levels near the ionization threshold.

The use of alternating kicks leads to a marked change in the pulse repetition frequency dependence of the Rydberg atom survival probability, which shows a general increase with increasing scaled frequency. Such behavior is expected. At high frequencies, the time delay between adjacent kicks becomes very short and the effect of one kick is, in essence, immediately reversed by the next (equal but opposite) kick. (Tests, shown in figure 5.11, in which the atoms were subject to 50 pairs of very closely spaced equal but opposite kicks showed that the survival probability was large and essentially independent of the pulse pair repetition frequency.) The data do, however, provide evidence for a local maximum in the survival probability for scaled frequencies
Figure 5.8. Poincaré surface of sections for the 1-D kicked atom for $\Delta p = -0.3$ and various $v_0$ values. The dashed line indicates the unperturbed torus associated with the initial state of the Rydberg atom.
Figure 5.9. Calculated survival probabilities for a 3D H(390p) atom subject to a train of (a) unidirectional and (b) alternating δ-function kicks with various scaled frequencies $v_o$ as a function of the number $N$ of kicks.

$v_o \sim 1$, and this is well reproduced by the CTMC simulations. This feature is suggestive of dynamical stabilization. To explore this possibility further, the dependence of the survival probability on $N$ was explored using the δ-function impulse model. These calculations (figure 5.9) indicate that, in contrast to the case of unidirectional kicks, the survival probability simply decreases monotonically with increasing $N$ for all scaled frequencies, even those in the vicinity of $v_o \sim 1$. (The rate of decrease, however, is significantly lower in this regime, leading to the local maximum in the observed survival probability.) Figure 5.9b suggests either that dynamical stabilization does not occur or that it occurs with very small probability. This result is, at first sight, surprising since the
survival probabilities after 50 unidirectional and bidirectional pulses have the same order of magnitude. This can be attributed to the fact that the chaotic diffusion rate is smaller for bidirectional than for unidirectional pulses. Only after a very large number of kicks does the effect of dynamical stabilization become evident and the survival probability for unidirectional pulses surpass that for alternating pulses. The presence of slow diffusion and absence of stabilization for alternating pulses is experimentally confirmed by the SFI profiles measured following application of 50 alternating HCPs (figure 5.10.c), which contain no pronounced feature at field strengths near those characteristic of parent-state ionization. Instead, the SFI spectrum is dominated by ionization of very-high-\(n\) atoms, pointing to diffusive motion toward higher \(n\) levels.

5.3 Probing the Localized State

Dynamical stabilization of Rb(390p) atoms subject to trains of unidirectional pulses was investigated further by examination of the final state of the atoms that remain after the HCP train using a half-cycle probe pulse. A schematic diagram of the HCPs applied appears in figure 5.12. Recall that application of a single HCP in the \(z\) direction delivers an impulsive momentum transfer \(\Delta p\) that will lead to an energy transfer of \(\Delta E = (\Delta p^2 /2) + p_{zi} \Delta p\) to the excited electron, where \(p_{zi}\) is its initial \(z\) component of momentum. For dynamical stabilization to occur, the energy transfer \(\Delta E\) from each successive HCP must (typically) be zero, which requires that the initial \(z\) component of electron momentum immediately before each HCP be \(p_{zi} \sim -\Delta p /2\). The \(z\) component of the electron momentum immediately after each HCP must then be \(p_{zi} \sim +\Delta p /2\) but must
Figure 5.10. SFI profiles of surviving atoms following application of 50 unidirectional HCPs with scaled pulse repetition frequencies $v_o$ of (a) 1.3 and (b) 0.5; and (c) following application of 50 alternating HCPs with $v_o = 1$. In each case the HCP amplitudes are sufficient to ionize ~90% of the parent atoms. The dashed line in each data set corresponds to the SFI profile observed for the parent Rb(390p) atoms. To better compare the shapes of the various profiles, the SFI spectra are normalized to equal areas.
Figure 5.11. Rydberg atom survival probability following application of 50 pairs of very closely spaced equal but opposite kicks.

Such evolution can be monitored using a probe pulse applied at various time delays after the HCP train whose amplitude is sufficient to ionize a significant fraction of the surviving atoms. Data obtained using this method are shown in figures 5.13 – 5.16. For each of these data sets, Rb(390p) atoms were subjected to a train of 20, 50, or 100 HCPs. The peak amplitudes of the pulses in the train were adjusted to achieve survival probabilities of 50% or 30%. A 500ps FWHM half-cycle probe pulse of 160mV/cm was applied after the last HCP in the train. The delay between the last HCP and the probe pulse was varied to examine the time evolution of $p_\tau$.

Figures 5.13 – 5.16 also include the results of CTMC simulations, which agree well with the data. Because the overall survival probability predicted by the simulations
was strongly dependent on the amplitude of the HCPs in the pulse train, the simulations were performed using the same method with which the experimental data were taken: the amplitudes of the pulse trains were adjusted for 50% and 30% survival probability and a 160mV/cm probe pulse was applied. The simulated field amplitudes agreed with the measured HCP field amplitudes to within 10%.

![HCP train diagram](image)

Figure 5.12. Schematic diagram of HCPs. The delay is measured between the last HCP in the train and the negative-going probe pulse.

As discussed in the previous section and shown in figure 5.8, dynamical stabilization occurs when the parent state overlaps with a stable island in phase space. Initial states that start on a stable island remain on that island. Those in the chaotic sea are eventually ionized. Thus, for pulse train frequencies leading to dynamical stabilization, most of the atoms that remain after the HCP train are localized in a small region of phase space. The values of $p_z$ for these localized states evolve coherently, leading to oscillations in the survival probability as a function of probe pulse delay. The
oscillations in the survival probability in figure 5.15 for \( v_o \sim 1.3 \) are therefore a signature of dynamical stabilization and of localization in phase space. The less pronounced oscillations for \( v_o \sim 0.6 \) indicate these processes are less effective. Because the localization takes place in an island that overlaps the parent state, the period of evolution of \( p_z \) is near the orbital period for the parent state and does not depend strongly on the pulse train frequency. For \( v_o \sim 0.7 \) and \( v_o \sim 3.0 \), no dynamical stabilization or phase space localization are expected. Indeed, figures 5.14 and 5.16 show no oscillatory behavior in the survival probability.

The beats damp because the HCP train populates a range of final \( n \) states, i.e., a range of energies. The smaller the range of \( n \) in the population that survives the pulse train, the slower the damping rate. Thus the \( p_z \) oscillations after 100 HCPs at \( v_o \sim 1.3 \) damp less rapidly than those after 50 or 20 HCPs.
Figure 5.13. Survival probability measured following application of a half-cycle probe pulse to Rb(390p) atoms that were subjected to a train of (a) 50 and (b) 100 unidirectional HCPs with $v_0 \sim 0.6$. The two data sets were taken with an initial Rydberg atom survival probability of $-50\%$ (●) and $30\%$ (○) following application of the HCP train. The solid line is the result of CTMC simulations for 50% survival probability.
Figure 5.14. Survival probability measured following application of a half-cycle probe pulse to Rb(390p) atoms that were subjected to a train of (a) 50 and (b) 100 unidirectional HCPs with \( v_o \sim 0.7 \). The two data sets were taken with an initial Rydberg atom survival probability of \(-50\% (\bullet)\) and \(30\% (\bigcirc)\) following application of the HCP train. The solid line is the result of CTMC simulations for 50\% survival probability.
Figure 5.15. Survival probability measured following application of a half-cycle probe pulse to Rb(390p) atoms that were subjected to a train of (a) 20, (b) 50, and (c) 100 unidirectional HCPs with $v_0 \sim 1.3$. The two data sets were taken with an initial Rydberg atom survival probability of $\sim 50\%$ (●) and $30\%$ (○) following application of the HCP train. The solid line is the result of CTMC simulations for 50% survival probability.
Figure 5.16. Survival probability measured following application of a half-cycle probe pulse to Rb(390p) atoms that were subjected to a train of (a) 50 and (b) 100 unidirectional HCPs with $v_0 \sim 3.0$. The two data sets were taken with an initial Rydberg atom survival probability of $\sim 50\%$ (●) and $30\%$ (○) following application of the HCP train. The solid line is the result of CTMC simulations for 50\% survival probability.
5.4 Response to Long Probe Pulses

The experimental investigation of the final states of Rydberg atoms subject to half-cycle pulses has relied on the use of very short HCPs to probe the \( z \)-component of the electron momentum \( p_z \). The first use of this technique was in probing the evolution of Rydberg wavepackets created by a single HCP. Here we discuss a complementary technique that provides information about the evolution of the electron spatial position. This provides a new tool to characterize the behavior and dynamics of coherently prepared Rydberg states.

Figure 5.17 shows the results of experiments in which a coherent state is prepared using a single HCP and probed by a second HCP of varying width. To produce the pulse sequences, the two outputs of the HP8131A pulse generator were combined using a matched power combiner. The relative delay of the pulses was stepped by computer control. The amplitude of the first pulse was 51 mV/cm \((\Delta p_o = 0.1)\) for all data sets. The amplitude of the second pulse was selected to achieve survival probabilities near 75%. For each probe pulse width, pronounced oscillations in the survival probability as a function of relative pulse delay are observed. As the width of the probe pulse is increased, the phase of the beats shifts and the amplitude decreases.
Figure 5.17. Rydberg atom survival probability following application of two HCPs as a function of relative time delay. The width of the probe pulse is indicated on each plot. The results for a 500ps probe pulse are included in each plot for reference.
Figure 5.17 continued.
Figure 5.17 continued.
Figure 5.18. CTMC simulations and experimental measurements of a high-\(n\) Rydberg atom subject to two HCPs as a function of relative time delay. The top two plots compare the expectation value of the \(z\) component of the electron momentum with the survival probability of the Rydberg atom probed with a short HCP. The bottom three plots compare \(<>\) with the survival probability for long probe pulses of two amplitudes.
The shifting stops when the probe pulse reaches a width of 20ns and the survival probability as a function of delay time has essentially the same form for 20ns, 30ns, and 50ns probe pulses. This is a surprising result because one would naively expect that for pulse widths greater than the classical orbital period the atoms would have plenty of time to respond to the field and the survival probability would therefore not vary with delay time. Rather than the snapshot of the atomic state provided by very short probe pulses, a long probe pulse would be expected to produce a response averaged over the length of the pulse. However, the beats remain. This behavior corresponds well to CTMC simulations using the measured experimental fields, shown in figure 5.18. The simulations indicate that the survival probability follows the beats of \( \langle p_z \rangle \) for short probe pulses and \( \langle z \rangle \) for long ones. Thus, long probe pulses provide a measure of the position of the electron.

Further interesting behavior is observed when the rise and fall time of the probe pulse is varied with its amplitude and width (FWHM) remaining constant. Figure 5.19 shows the measured survival probabilities as a function of relative pulse delay for 20ns probe pulses with an amplitude of 28mV/cm. The oscillations in survival probability decrease in amplitude as the rise time increases and disappear by a rise time of 10ns. Clearly, a rise time that is short compared to the classical orbital period of the parent state is necessary to probe the electron position. The rise time must be short to produce coherent excitation to the final state, thus producing the pronounced variations in electron position observed.
Figure 5.19. Measured survival probability following application of two HCPs as a function of relative time delay. The second HCP has a FWHM of 20ns and an amplitude of 28mV/cm. The rise time of the second HCP is indicated on each plot.
Chapter 6

Conclusions

6.1 Conclusions

High-\(n\) Rydberg atoms provide an excellent vehicle to investigate the response of atoms to pulsed electric fields and can be used to study impulsively-driven systems, classical quantum correspondence, wavepacket dynamics, and the control and manipulation of atomic wavefunctions. High-\(\ell\) states and phase-space localized states can also be created and probed using HCPs. In addition, HCPs provide a means to examine both the momentum and spatial distributions of product states. The techniques developed in the present work may be used in further studies described in the following section.

6.2 Future Work

High-\(n\) Rydberg atoms provide the basis for a wide range of future experiments. Extensions of the present work include studies of impulsively driven atoms and ultra-low energy electron/molecule scattering.

6.2.1 Dynamical Stabilization

The character of the dynamical stabilization described in chapter 5 for Rydberg atoms subject to trains of HCPs seems to depend on the average value of the electric field over the pulse train. Applying an offset to the pulses as shown schematically in figure 6.1 could alter the stabilization and elucidate the importance of an average DC field in enabling dynamical stabilization, even for trains of alternating HCPs.
Preliminary CTMC simulations suggest that the fraction of atoms that undergo dynamical stabilization will be strongly dependent on the initial electronic state. The unperturbed atomic Hamiltonian $H_{at}$ is given in cylindrical coordinates $(z, \rho, \phi)$ by

$$H_{at} = \frac{p_z^2}{2} + \frac{p_\rho^2}{2\rho^2} + V_{at}\left(\frac{\sqrt{z^2 + \rho^2}}{\rho^2}\right)$$  \hspace{1cm} (6.1)$$

where $L_z$ is the $z$-component of angular momentum, which is a constant of the motion, and $V_{at}$ is the potential due to the core ion. For the $np$ states studied to date, $L_z$ is very small. Theory suggests, however, that the for large values of $L_z$ the increased centrifugal barrier ($L_z^2/2\rho^2$) will increase the likelihood of dynamical stabilization, which might even be observed for alternating kicks. This can be investigated by studying the response of high-$\ell$, high-$m$ atoms to a variety of HCP trains. High-$\ell$ states can be generated using an applied electric field of carefully controlled duration and amplitude as shown in chapter 5.1. To generate high-$m$ states, the field must be applied perpendicular to the $z$ axis.

The role of noise in suppressing dynamical stabilization might also be investigated. Experiments with Rydberg atoms ionized by coherent microwave fields
have shown that dynamical localization is destroyed by noise. Preliminary 1D simulations for short, unidirectional kicks indicate that noise superposed on the HCP train leads to a reduction in the overall survival probability and to a narrowing of the frequency intervals over which dynamical stabilization occurs.

6.2.2 Intense Field Stabilization

When an atom interacts with a radiation field, the probability of ionization is a function of the initial state, the interaction time, the field intensity, and the field frequency. One would expect that a stronger field would always produce a larger ionization. However, theoretical investigations have shown that when the field is increased above a certain critical value, a hydrogen atom becomes more stable against ionization. This effect is known as intense field stabilization (IFS).

Casati presents a model of a classical 3D hydrogen atom subject to a periodic sequence of δ pulses of fixed strength that alternate in sign. The Hamiltonian for this system becomes

$$H(p,r,t) = \frac{p^2}{2} - \frac{1}{r} + \frac{2\varepsilon}{\omega} \sum_{n=-\infty}^{\infty} (-1)^n \delta \left( t - \frac{nT}{2} \right)$$

(6.2)

where $\varepsilon$ is the intensity and $\omega$ the frequency of the δ pulses, so that $\Delta p_z = \pm 2\varepsilon/\omega$. As shown in figure 6.2, their results indicate that the threshold for IFS is $\varepsilon = 5\omega/m$. The calculations suggest that IFS is suppressed when the kicks are simply turned on at their full amplitude. Instead, they must increase in amplitude from zero to their peak value over several kicks.

6.2.3 High Scaled Frequencies
Although our measurements to date can be well explained using either classical or quantum physics, the correspondence between classical and quantum dynamics is expected to break down at some point, even in the limit of very large $n$. Recent 1D calculations suggest that such a breakdown might become evident at high scaled frequencies, $v_o \geq 15$. Figure 6.3 shows survival probabilities calculated using classical and quantum models for an $n=50$ Rydberg atom as a function of the number $N$ of kicks. Initially the classical and quantum predictions mirror each other and exhibit a

![Graph](image)

Figure 6.2. (from reference 42) Survival probability after a time ~1000 a.u. of application of $\delta$-function pulses of intensity $\epsilon$ and frequency $\omega$. Darker tones of gray correspond to larger survival probability. The line has equation $\epsilon = 5\omega m$. 
pronounced N-dependence in the survival probability. For N > 30 kicks, however, the predictions begin to diverge, which could be indicative of quantum suppression of classical chaos. Studies at such high scaled frequencies will require the use of states of much higher n. Selective single-photon excitation of Rb states with n>400 is difficult due to the overlapping spectra of the $^{87}\text{Rb}$ and $^{85}\text{Rb}$ isotopes and the large hyperfine ground-state splitting. However, these states may be reached in potassium, which has a much smaller ground state hyperfine splitting, or using

![Graph showing 1D H(n=50) kicked atom for a scaled frequency of 16.8](image)

**Figure 6.3.** Quantum and classical calculations of the survival probability vs the number of $\delta$-function kicks for a scaled frequency of 16.8.

multiple transitions in rubidium.
6.2.4 Ultra-Low-Energy Electron/Molecule Scattering

In collisions with neutral targets, a high-n Rydberg atom behaves not as an atom but as a pair of independent particles. Studies of Rydberg atom collision processes that result from an electron/target interaction can provide information on electron/molecule scattering at energies extending down to a few μeV. At high-n, collisions with polar targets frequently leads to ionization through transfer of molecular rotational energy to the excited electron in a dipole-allowed $J \rightarrow J-1$ rotational transition. The rate constant associated with such collisional ionization can be written

$$k = \int_0^\infty v \sigma(v) f(v) dv \quad (6.3)$$

where $f(v)$ is the electron velocity distribution and $\sigma(v)$ the cross section for superelastic electron scattering, which is averaged over the target rotational distribution. Application of a long electric field pulse to high-n atoms results in periodic variations in $f(v)$. The presence of dipole-supported bound or virtual states introduces resonances in the cross section for superelastic scattering, which takes the form

$$\sigma(v) = \frac{1}{v^{2-x} (v^2 + \kappa^{2x})} \quad (6.4)$$

where $-\kappa^2/2$ is the energy of the dipole-supported state and $x$ is a parameter related to the non-averaged component of target dipole moment. Given a cross section of this form, periodic changes in $f(v)$ will lead to periodic changes in $k$ and the ionization rate whose size depends on the location of the dipole-supported resonance within the (evolving) electron velocity distribution. These changes in $k$ are estimated in figure 6.4 for $x=1$. 
Figure 6.4. Estimated ratio of the rate constant for collisional ionization for $np$ states to that for $\ell$-mixed states as a function of $\kappa$ where $E = -\kappa^2/2$ is the energy of a dipole-supported state.
References


