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IONIZATION OF RYDBERG ATOMS BY HALF-CYCLE PULSES:
EFFECT OF PULSE SHAPE AND RISE TIME

by
BRIDGET E. TANNIAN

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J. Young, Director
Professor of Electrical and Computer
Engineering

F. B. Dunning,
Professor of Physics

F. K. Tittel,
Professor of Electrical and Computer
Engineering

Houston, Texas

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IONIZATION OF RYDBERG ATOMS BY HALF-CYCLE PULSES: EFFECT OF PULSE SHAPE AND RISE TIME

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Abstract

The ionization of potassium np Rydberg atoms with $n \sim 388$ by pulsed unidirectional electric fields, termed half-cycle pulses (HCPs), with various well-characterized shapes (rectangular, triangular, and sawtooth) and durations T_p comparable to the classical electron orbital period T_n is investigated. The experimental results are compared to classical trajectory Monte Carlo (CTMC) simulations and the classically-scaled results of quantum calculations undertaken at $n=5$. The data show that in the intermediate regime, $T_p \sim T_n$, the threshold field for ionization becomes sensitive to the shape and rise time of the HCP, increasing with increasing rise time. For each pulse shape, the agreement between the experimental measurements and both the CTMC and quantum calculations is very good.

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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 a Rydberg atom resemble those of hydrogen. The orbital period 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 $n \sim 400$, rising to >100 ns at $n \sim 1000$, a time scale that is easily observed on a laboratory oscilloscope.

Table 1.1 Properties of Rydberg Atoms

Property	scaling (a.u.)	$n=1$	$n=30$	$n=400$	$n=1000$
Mean radius	n^2	5.3×10^{-9} cm	4.8×10^{-6} cm	8.5×10^{-4} cm	5.3×10^{-3} cm
Orbital Period	$2\pi n^3$	1.5×10^{-16} s	4.1 ps	9.6 ns	150 ns
Binding Energy	$\frac{1}{2n^2}$	13.6 eV	15 meV	85 μ eV	13.6 μ eV
Energy Spacings	$\frac{1}{n^3}$	10.2 eV	1.0 meV	0.43 μ eV	27.2 neV
Classical Field Ionization Threshold	$\frac{1}{16n^4}$	3×10^8 Vcm $^{-1}$	400 Vcm $^{-1}$	13 mVcm $^{-1}$	320 μ Vcm $^{-1}$

The response of atoms in high-lying Rydberg states to unidirectional pulsed electric fields, termed half-cycle pulses (HCPs), has been investigated previously in this laboratory^{1,2} and elsewhere.^{3,4} These investigations show that the field amplitude required for ionization of highly excited Rydberg atoms by unidirectional pulsed electric fields depends critically on the pulse duration. For pulses much shorter than the classical orbital period, ionization results when the impulse delivered to the excited electron is sufficient to increase its energy by an amount that is greater than its original binding energy. For long duration pulses, ionization results primarily from tunneling through the potential barrier formed by the supersposition of the atomic potential and the external field or from over-barrier escape. The transition from the short pulse to the long pulse regime is of particular interest.

Although the previous investigations all showed that the threshold field for ionization depends on pulse duration, the results for the values of those threshold fields disagreed, as shown in figure 1.1. Kristensen *et. al.* concluded that the classical scaling techniques used to produce the values in figure 1 were not applicable to the values of n used in their experiments, $n \sim 40$, because the classical limit is not reached for such n . However, the shapes of the field pulses differed between experiments. In this work, we investigate whether the discrepancies can be explained in terms of the effects of pulse shape.

We have measured the ionization probabilities for potassium Rydberg atoms with $n \sim 400$ subjected to HCPs of several shapes. For such high n , the classical electron orbital period is quite long, allowing ionization and excitation to be investigated, even in the short-pulse regime, by using nanosecond pulses from conventional fast pulse generators. These pulses can be accurately characterized so that comparison can be made to theory on an absolute scale without any adjustable parameters. In contrast, to perform the same experiment at $n=30$, one would need to produce pulses around 1ps in width. Such pulses would be far more difficult to produce and characterize. This work will show that in the

transition regime, accurate characterization of applied pulses is required to predict ionization thresholds and that classical scaling is appropriate for this and previous experiments.

Figure 1.1. Scaled threshold fields $F_o = n^4 F$ (a.u.) for 10% ionization as a function of the scaled pulse duration $T_o = T_p/T_n$. The results of Frey et al (○) (rectangular pulses) and Kristensen et al. (◆) (Gaussian pulses) do not agree. The results of Jones et al. (△) for short pulses are shown. Classical trajectory Monte Carlo calculations for idealized rectangular (—) pulses are included.

Chapter 2

Field Ionization

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 has one electron bound to one proton by the Coulomb potential, $V(r) = -1/r$, where r is the distance between the charges. The classical Hamiltonian is therefore $H = \vec{p}^2 - 1/r$.⁵ 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 $E < 0$ 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 by⁶ $e = \sqrt{1 + 2EL^2}$. 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

$$x = a(\cos \zeta + e), \quad y = a\sqrt{1 - e^2} \sin \zeta$$

where

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, $L_0 = L / \sqrt{2|E|}$.

$$t = \sqrt{a^3} (\zeta + e \sin \zeta)$$

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

$$T = 2\pi a^{3/2} = \pi \sqrt{2|E|^{-3}}.$$

The momentum of the electron is

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

with

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

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 $p = p_{rms} = \sqrt{2|E|}$.

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

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.¹

The large number of states that must be considered when $n \geq 200$ makes quantum mechanical calculations difficult.⁷ 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 set⁸

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

where the Sturmian parameter $\lambda = 1/n_s$ defines the set. States with $1 \leq n \leq n_{\max}$ are used, which, for $m=0$, corresponds to a number of states $N_{\max} = n_{\max}(n_{\max} + 1) / 2$. The parameters λ and $N_{\max}$ 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 $\vec{F} = F(\hat{z})$ is⁹

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

The above equation is separable in parabolic coordinates.

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

Defining the wave function Ψ as the product $\Psi = (\xi\eta)^{-1/2} V(\xi)U(\eta)e^{\pm im\varphi}$, we obtain

$$\begin{aligned}\frac{d^2V}{d\xi^2} + \left(\frac{E}{2} + \frac{\beta_1}{\xi} + \frac{1-m^2}{4\xi^2} - \frac{F}{4}\xi \right) V &= 0 \\ \frac{d^2U}{d\eta^2} + \left(\frac{E}{2} + \frac{\beta_2}{\eta} + \frac{1-m^2}{4\eta^2} - \frac{F}{4}\eta \right) U &= 0\end{aligned}$$

where β_1 and β_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

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

where n_1 and n_2 are parabolic quantum numbers with $n = n_1 + n_2 + |m| + 1$. Figure 2.2 is a plot of the H $|m| = 1$ energy levels from $n=8$ to $n=14$ in electric fields up to 10^5 Vcm⁻¹, strong enough to field ionize the atom. Note that the levels shift approximately linearly with F up to field strengths sufficient to ionize the atom. Also, notice 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 2.3 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

$$z_s = -\sqrt{\frac{1}{F}}, \quad V_s = -2\sqrt{F}.$$

Figure 2.2 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 Gallagher).

Figure 2.3. Potential energy for a one-electron atom in an electric field of $f = 0.02$ 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 2.4 [from reference 10] shows the charge distribution for hydrogen for parabolic eigenstates ($k=n_1-n_2$). The lower energy states are localized close to the Stark saddle and will ionize near the classical threshold. The higher energy states are localized away from the saddle point and ionize at higher fields.

Because the potential falls away in the $-z$ direction (see figure 2.3), there are no true stationary states for an atom in an electric field.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.

Figure 2.4. Electron spatial probability distribution for hydrogen for $n=8$ and $m=0$ Stark states. n_I varies from 7 to 0.

2.3 Non-hydrogenic Orbits

The Hamiltonian in equation 1.1 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

$$E_{n\ell} = -\frac{1}{2(n - \delta_\ell)^2}$$

where the quantum defect δ_ℓ 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 ℓ . 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.

$$\Delta E = 2 \left| \langle n, n_1, n_2, m | V_d(r) | n', n'_1, n'_2, m' \rangle \right|$$

where V_d is the difference between the real potential and a pure Coulomb potential.¹¹ For low m , $|m| \ll n$, the energy separation at avoided crossings between Stark states from neighboring n manifolds scales approximately as $1/n^4$. States of low ℓ have a lot of core interaction and therefore exhibit larger energy gaps at avoided crossings than states of high ℓ .¹² The calculated energy levels of potassium Rydberg states near $n \sim 100$ as a function of field strength are shown in figures 2.5 and 2.6 (fig 2.11 and 2.12 from Mark). Note that the Stark states do not cross.

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 2.7.

The probability of traversing an avoided crossing diabatically is given by the Landau-Zener formula¹³

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

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 2.5. Potassium $lml = 0$ Stark levels near $n=100$. (from reference 1)

Figure 2.6. Potassium $lml = 1$ Stark levels near $n=100$. (from reference 1)

Figure 2.7. Adiabatic or diabatic passage of state B through an avoided crossing with state A.

Chapter 3

Experimental Apparatus

In the present work, potassium atoms in a collimated beam are photoexcited by a frequency doubled tunable ring dye laser. Excitation occurs near the center of an interaction region, shown in figure 3.1. The interaction region is defined by three pairs of planar copper electrodes that are independently biasable in order to minimize stray electric fields. The laser output is chopped into a train of pulses and measurements are conducted in pulsed mode. Following each laser pulse, the Rydberg atoms are exposed to a half-cycle pulse (HCP), which is generated by applying a voltage pulse to a small circular copper disc inset in 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.

3.1 Vacuum System

The vacuum system has two chambers with a 0.5mm aperture in between. The chamber containing the potassium oven is pumped by a 4" diffusion pump. The main chamber contains the interaction region in which 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 $\sim 1 \times 10^{-7}$ torr. The laser beam enters and exits the main chamber through quartz windows set at Brewster's angle.

3.2 Potassium Beam

The potassium oven is an enclosed stainless steel reservoir with a 0.5mm hole. The reservoir is heated to 300°C and the exit hole to 340°C by resistance cartridge heaters monitored by thermocouples and controlled by microprocessor temperature controllers. The exit hole is set at a higher temperature to prevent potassium from condensing on it. The potassium beam passes through a 0.5mm collimating aperture into the main chamber, resulting in a beam divergence of ≤ 1 degree. The beam density is estimated to be $\sim 10^9 \text{cm}^{-3}$ at the center of the interaction region.¹⁴ The average velocity of the potassium atoms is $\sim 570 \text{ m/s}$.

3.3 Laser System

The wavelength needed to excite $n \sim 388$ potassium Rydberg atoms by single photon excitation is 2856.40 Å. This is provided by an intercavity frequency doubled ring dye laser, a Coherent model CR-699-21 with a model 7500-06 doubling package, which 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.¹⁵ 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 by second harmonic generation, producing 1-4 mW of UV output.

Figure 3.1. Schematic diagram of the apparatus.

The laser frequency is stabilized by a temperature controlled Fabry-Perot etalon. Deviations in frequency from the lock point are corrected by changing the optical length of the laser cavity. Fast deviations are corrected using a PZT mounted mirror and slow ones by rotating a 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 helium-neon laser using another temperature controlled Fabry-Perot etalon.¹⁶ The etalon mirror spacing is scanned at 50 Hz, producing two series of transmission peaks, one for each wavelength. Changes in the separation of a pair of peaks are used to generate error signals to restore the laser to the lock

point. The overall frequency stability of the dye laser is $\sim \pm 1$ 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. The accuracy of the wavemeter is ± 1 GHz at the operating frequency.

The UV output of the dye laser is chopped into $4\mu\text{s}$ 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 potassium beam. It is surrounded by three pairs of copper electrodes, each 10 cm x 10 cm (see fig. 3.1). Electric field pulses in the interaction region are created by applying voltage pulses to a circular copper disk, 4 cm in diameter, inset into the top plate. This electrode is terminated by $50\ \Omega$ to ground. To reduce RF pickup on the other electrodes, the top plate is grounded, the HCP electrode is shielded, and RF-decoupling capacitors are connected between the side plates and ground.

Rydberg atoms remaining in the interaction region after the HCP are ionized in the electric field generated by a slow voltage ramp applied to the bottom plate. Electrons from the ionized Rydberg atoms are accelerated by the extraction ramp through a mesh-covered hole in the bottom plate into a Johnson Laboratory MM-1 electron multiplier.

DC electric fields in the interaction region are reduced to $\leq 50 \mu\text{V}/\text{cm}$ by the technique described in section 4.1. Magnetic fields are reduced to $\sim 20 \text{ mG}$ by a μ -metal shield that surrounds the interaction region.

3.5 Half-Cycle Pulses

A Hewlett Packard 8082A pulse generator is used to produce the half-cycle pulses (HCPs). It can generate pulses with amplitudes up to $\sim 5 \text{ V}$. The risetime and fall time of the pulses are independently adjustable for values greater than 5 ns and can be adjusted together to $\sim 1 \text{ ns}$. The amplitudes of the pulses are adjusted using broadband (DC - 2 GHz) attenuators in series with the pulse generator. In order to supply both a DC bias and an 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.

3.6 Data Acquisition System

Data are accumulated over many identical cycles under computer control. The master pulse generator produces pulses at $\sim 10 \text{ kHz}$ to begin the timing sequence shown in figure 3.2. Figure 3.3 shows the setup of the data acquisition system.

Figure 3.2. 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 3.3. Block diagram of the data acquisition system.

The master pulse generator produces logic pulses to synchronize other electronics and timing devices at a repetition rate of ~ 10 kHz. Each pulse from the master pulse generator begins the timing sequence shown in fig. 3.2. One output pulse triggers the acousto-optic modulator to allow a laser pulse of ~ 4 μ s duration to enter the apparatus and produce Rydberg atoms. Another pulse from the master pulser triggers a computer-controlled gate (CCG) which, when enabled by the computer, in turn triggers a SRS DG535 precision computer-controlled delay generator. The DG535 dual outputs trigger the HCP pulse generators that control the time separation between the two HCPs to better than 50 ps. A third pulse from the master pulser triggers an Evans 4145 programmable delay module. The Evans delay module triggers the SFI voltage ramp and simultaneously starts the time-to-amplitude converter (TAC) at a selected delay time, typically a few μ s. Electrons resulting from field ionization are swept out of the interaction region by the voltage ramp and are detected by the MM-1 electron multiplier. Output pulses from the MM-1 are amplified and compared to a preset threshold by a LeCroy MVL100 low level amplifier/comparator. The first output pulse of the MVL100 is used to stop the TAC (the probability that a Rydberg atom is produced in any one laser pulse is low, ≤ 0.05). The TAC produces an output pulse whose height ranges between 0 and 10 V and is proportional to the time difference between the start and stop pulses, i.e. to the time between the start of the voltage ramp and the time the electron arrives at the detector. Because atoms in different Rydberg states ionize at different applied fields, measurement of the field ionization signal as a function of electron arrival time, i.e. of applied field, provides a measure of the excited-state distribution of those atoms surviving the HCPs (see section 4.1). The TAC output amplitude is digitized by a 12-bit analog-to-digital converter (ADC) and recorded by the computer. The computer disables the the CCG so that no HCPs are applied in alternate sets of experimental cycles to monitor the rate of production

of Rydberg atoms. The Rydberg atom survival probability is obtained by comparing the Rydberg atom signals observed with and without HCPs present.

The ADC and the digital lines that control the CCG and Evans delay module are located on a National Instruments LAB-NB NuBus multifunction data acquisition board plugged into an Apple MacIntosh Quadra 800. The LAB-NB also has two 12-bit digital-to-analog converters (DACs) which can each produce analog voltages from -5V to +5V. These analog voltages are used as control voltages to scan the laser frequency or the potential difference across an interaction region electrode pair .

The SRS DG535 delay generator is interfaced to the computer by a General Purpose Instrument Bus (GPIB) *via* a GPIB controller.

Chapter 4

Experimental Techniques

4.1 Control of Stray Electric Fields

The presence of a stray electric field can reduce the rate of Rydberg atom excitation. Figure 4.1 shows the effect of small electric fields on the excitation of Rydberg states around $n \sim 270$. Any stray electric field in the interaction region will cause Stark mixing of np states with the neighboring $n\ell$ manifold. Thus, the state excited by the laser is not a well-defined p state, resulting in diffusion of the $4s \rightarrow np$ oscillator strength and a lower excitation probability for a given laser frequency.¹² A field strength of $\sim 100 \mu\text{V/cm}$ will reduce the excitation probability at $n \sim 400$ by half.

A technique has been developed to reduce these stray electric fields using the quadratic Stark shift.¹⁷ The laser is detuned a few MHz from a $4s \rightarrow np$ transition. The bias applied between a pair of copper electrodes is ramped slowly and the Rydberg atom production recorded. Figure 4.2(a) shows the Rydberg atom production as a function of applied electric field. The midpoint between the peaks is determined by minimizing the quantity

$$\varepsilon(E_0) = \sum_n |S_{E_0 + n\Delta E} - S_{E_0 - n\Delta E}|, \quad (4.1)$$

where $S_{E_0 \pm n\Delta E}$ are the Rydberg atom signals observed with the applied fields $E_0 \pm n\Delta E$, and the selected offsets $n\Delta E$ encompass the observed peaks. Applying the field corresponding to the midpoint reduces the electric field in that direction to less than ± 30

Figure 4.1. Effect of an applied electric field on the excitation of potassium Rydberg states in the vicinity of $n \sim 270$ with (a) $m = 0$ and (b) $|m| = 1$.

Figure 4.2. Rydberg atom signal observed as a function of applied electric field for excitation of Stark states with $n=150$ and $n=250$. (b) Dependence of $\epsilon(E_0)$ (see eq.4.4) on the assumed value of E_0 . The midpoint between the peaks is denoted by E_{0m} .

$\mu\text{V}/\text{cm}$. After using the procedure in each direction, the total residual field in the interaction region is $\leq 50 \mu\text{V}/\text{cm}$.

The field zeroing procedure is slightly different in the y direction (as defined by figure 3.1). The y direction is parallel to the polarization of the laser beam, so the Rydberg states in the $m=0$ Stark manifold will be excited. Figure 4.1 shows that, in an electric field, the largest oscillator strength for $m=0$ is for the "s-like" Stark state. Therefore, this state is used, as it also has a quadratic dependence on field.

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 to the interaction region. If there is a Rydberg atom in the interaction region, it will field ionize. Because different states ionize in different fields, the time dependence of the ionization signal gives an indication of the initial state of the Rydberg atom. Thus, SFI is useful both to detect the presence of Rydberg atoms and to determine the excited state distribution.

The physics of ionization by ramped electric fields is discussed in chapter 2 and treated in more detail elsewhere.^{10,11,18} In small fields, less than the Stark crossing field of $\sim 1/3n^5$, the 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 . In the present work, the frequency separations between adjacent Stark states with $n \sim 400$ 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 $F \sim 1/9n^4$. 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 the HCP will also be extracted by the field ramp and can contaminate the SFI spectrum. Thus there must be a delay between 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 $\leq 5 \mu\text{s}$.¹ For the present work, delay times of $7 \mu\text{s}$ were used.

Chapter 5

Results and Discussion

The survival probabilities of Rydberg atoms subject to rectangular half-cycle pulses have been previously measured in this laboratory.^{1,2,7} In those data, the ionization probabilities satisfy classical scaling invariance and display the predicted crossover from a $1/n^4$ to a $1/n$ scaling in threshold fields as the pulse duration changes from the adiabatic to the sudden limit. (see figure 1.1) Recently, Kristensen et al.⁴ found threshold fields for Gaussian pulses different from those in reference 2 and attributed the difference to a failure of classical scaling invariance. This work is an attempt to determine whether the difference is due to the effect of pulse shape on ionization threshold.

5.1 Description of the HCP Pulse Shape Experiment

Electric field pulses of several durations and several shapes were applied to $n \sim 388$ potassium Rydberg atoms.^{8,19} Figure 5.1 shows the HCP shapes. The pulse durations, defined as the full width at half maximum pulse height, vary from 5ns to 50ns. The classical orbital period for an electron at $n \sim 388$ is about 9ns, so the pulse durations go from the short-pulse limit to the long-pulse limit. The four pulse shapes are rectangular (rect), fast-rise/slow-fall (frsf), triangular (srsf), and slow-rise/fast-fall (srff). Numerical integration of the measured pulse profiles shows that, for each value of peak height and FWHM, the pulse areas are equal to within $\pm 10\%$. The area of the rectangular pulse is typically the smallest.

The survival probability is the ratio of the number of Rydberg atoms surviving with no HCP to the number surviving after an HCP is applied.

Figure 5.1 Measured HCP shapes for 5ns, 10ns, 15ns, 20ns, 30ns, 40ns, 50ns, and 100ns pulse durations. From the bottom of each set, pulses are rectangular, frsf, srsf, and srff. Missing pulses could not be produced.

5.2 Results

Figure 5.2 shows the survival probability as a function of pulse height for each pulse shape for 10ns and 40ns pulses. The data agree well with the predictions of CTMC theory, which are also included in the figure. Note that the shape of the survival probability curve is different for each HCP shape. The difference is less pronounced for short pulse durations. Also, the frsf data lie almost on top of the rectangular pulse data, indicating that the ionization probabilities are not sensitive to the fall time of the pulse.

The threshold fields for 10% ionization are shown in figure 5.3 as a function of pulse duration scaled by the classical orbital period. The data of Frey *et al.* data using rectangular pulses and that of Kristensen *et al.* using Gaussian pulses are also included. Theoretical predictions produced using a finite Sturmian basis set at $n=5$ and classically scaled agree well with the data. These threshold fields were calculated for an ideal rectangular pulse and an ideal srsf triangular pulse. Unfortunately, results for $n>5$ or $T_0 \geq 4$ could not be obtained. However, the agreement between theoretical prediction and data suggests that even at $n=5$ the scaling invariant limit, where classical scaling is appropriate, is being approached. Work on extending calculations to larger T_0 is in progress.

5.3 Discussion

The dependence of threshold field on pulse shape changes as the pulse duration goes from short to long. For a pulse that is long compared to the classical orbital period, ionization comes about through a modification of the electronic binding potential, as in the Stark effect described in chapter 2. If the pulse is short, the electron does not have time to respond to the rapid changes in the potential. It can only ionize by gaining energy from the field. Thus, pulse shape has little effect on ionization threshold for short pulses. The probability for ionization by a short pulse depends only on the impulse $\Delta p = -\int F(t)dt$ delivered by the field $F(t)$ to the excited atom, which is essentially the same for each pulse

shape of a given duration. Note that in figure 5.2, the ionization probability depends less strongly on pulse shape for the 10ns ($T_o \approx 1$) pulses than for the 40 ns ($T_o > 1$) pulses. Also, in figure 5.3, the threshold fields for different pulse shapes converge as T_o gets shorter.

For long pulses, the path taken to ionization depends on HCP rise time. However, the frequency separations of between adjacent Stark states of $n \sim 400$ at the avoided crossings are much less than the rate at which the applied ramp rises.¹ Thus, the avoided crossings between neighboring Stark states described in chapter 2 do not play a large role in the observed dependence of the ionization threshold on rise time because they are all traversed diabatically for any of the pulse shapes used. A much longer rise time would be needed for the electron to follow an adiabatic path to ionization. Previous experiments in this laboratory¹⁸ showed that for the sodium $34d$ state, the path to ionization was entirely diabatic for slew rates above 10 V/cm $\cdot\mu$ sec. Since the energy spacing in avoided crossings goes as $1/n^4$, as discussed in chapter 2, for the $n=400$ state of sodium, the slew rate required for diabatic passage would be approximately 7×10^{-5} V/cm $\cdot$ ns. For a pulse with a rise time of 100ns, the longest rise time used in this work, and a peak amplitude of 50mV/cm, the slew rate is 5×10^{-4} V/cm $\cdot$ ns, which is certainly fast enough for entirely diabatic ionization.

A proposed explanation for the effect of rise time on ionization probability is that near the classical ionization limit, the electron encounters a "hidden crossing" in the potential.^{20,21} Theoretical calculations show that for ion-atom collisions, there is an avoided crossing between the red-most state of the n Stark manifold and the red-most state of the $n+1$ manifold. In an ion-atom collision, the potential seen by an atomic electron is the atomic potential plus a time-dependent potential due to the ion moving by. This situation is similar to our experiment with an electric field pulse, and such avoided crossings may also occur there. These avoided crossings have about the same energy

separation as is between the zero-field n and $n+1$ states. This energy separation corresponds in frequency to a few classical orbital periods. A fast-rising pulse would cause the electron to traverse this "hidden crossing" diabatically. If the pulse has a rise time longer than the classical orbital period, there is a greater probability of traversing the "hidden crossing" adiabatically. The atoms with a diabatic passage will reach the ionization threshold sooner and will ionize at a faster rate than those with an adiabatic passage. Pulses with faster rise times will have lower ionization thresholds than those with slower rise times.

Figure 5.2. Measured and calculated (CTMC) survival probabilities as a function of HCP amplitude expressed in scaled units $F_0 = n^4 F$ (a.u.) for the ($\square$, ———) rectangular, (Δ , ———) srsf triangular, (∇ , ———) frsf triangular, and (O , ———) srff triangular pulses and pulse durations of (a) 10ns and (b) 40ns.

Figure 5.3. Scaled threshold fields $F_0 = n^4 F$ (a.u.) for 10% ionization as a function of the scale pulse duration $T_0 = T_p/T_n$. The symbols used for the various experimental data are as in figure 5.2. Classically-scaled quantum calculations for idealized rectangular (—) and triangular (— —) pulses are included. The earlier results of Frey et al (■) (rectangular pulses) and Kristensen et al. (◆) (Gaussian pulses) are included for comparison.

Chapter 6

Conclusions

The present work demonstrates that when the HCP duration becomes comparable to or greater than the electron orbital period, the ionization threshold becomes sensitive to rise time. Therefore, accurate HCP characterization is required to predict ionization thresholds.

Studies of ionization of potassium Rydberg atoms using HCPs with durations that span the transition from the short-pulse to the long pulse regime show that at very-high- n the classical limit of ionization is reached and quantum corrections are negligible. CTMC theory and quantum mechanical theory are able to accurately predict absolute ionization and survival probabilities.

This work can be extended using alternating pulses of different rise and fall times. These pulses would be like one cycle of an RF field. There would be no impulse delivered to the system, but if the RF frequency were close to the classical orbital frequency, the field direction would follow the direction of the motion of the electron, perhaps resulting in ionization. Studies of ionization by a sequence of HCPs whose directions alternate may be used to test theories of intense field stabilization. Koch and van Leeuwen²² discuss several regimes of microwave frequency in ionization studies. In the regime where the microwave frequency is greater than the classical orbital frequency, there is always a threshold amplitude for ionization below which the atom does not ionize and above which it does. However, it has been predicted that for very intense fields, the atom will not ionize. This phenomenon is referred to as intense field stabilization. Calculations performed for a one-dimensional model atom²³ show that an electric field strength $E_0=5.0$ is sufficient for stabilization. To perform an intense field stabilization experiment with atoms at $n=30$,

fields of thousands of $\text{V}\cdot\text{cm}^{-1}$ would be necessary. However, at $n\sim 400$, only tens of $\text{mV}\cdot\text{cm}^{-1}$ would be needed.

There are many other experimental studies of interest that can be performed with HCPs and very-high- n Rydberg atoms. The dynamics of wavepackets created by HCPs in static electric and magnetic fields can be studied. For these systems, the underlying classical dynamics can be chaotic.²⁴

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